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(54) Title: ULTRA-LOW MOLECULAR WEIGHT ETHYLENE POLYMERS

(57) Abstract

The subject invention pertains to non-pourable homogeneous ultra-low molecular weight ethylene polymers and a process for the preparation thereof. Such polymers have longer lamella and greater degree of crystalline organization than corresponding higher molecular weight materials at an equivalent density.

5   Ultra-Low Molecular Weight Ethylene Polymers

The subject invention pertains to ethylene polymers having an ultra-low molecular weight, as evidenced by a low number average molecular weight. In particular, the subject invention pertains to ethylene polymers having a number average molecular weight as

10   determined by gel permeation chromatography of no more than 11,000.

U.S. Patent No. 3,645,992 discloses homogeneous linear ethylene olefin copolymers prepared using a soluble vanadium catalyst. Therein, homogeneous copolymers are defined as polymers in which the comonomer is randomly distributed within a given molecule, and in which all copolymer molecules have the same ethylene to copolymer ratio. The disclosed

15   homogeneous copolymers have a moderately high molecular weight. For instance, as set forth in the Examples, the homogeneous copolymers have a melt index, as measured in accordance with ASTM D-1238, of from less than 0.1 to less than 25 g/10 min.

U.S. Patent Nos. 5,272,236 and 5,278,272 disclose substantially linear ethylene olefin copolymers prepared using a single site polymerization catalyst. The disclosed substantially

20   linear copolymers are characterized as having from about 0.01 to about 3 long chain branches per 1000 carbons. Unlike the homogeneous copolymers of Elston, the disclosed substantially linear copolymers are characterized by a molecular weight distribution (M_w/M_n) which is independent of the I_{10}/I_2, as measured in accordance with ASTM D-1238.

Pourable ultra-low molecular weight ethylene polymers for use as oil additives are

25   known in the art. For instance, PCT published application 94/12193 discloses ethylene/butene copolymers having a number average molecular weight between 1500 and 7500 prepared using a biscyclopentadienyl metallocene catalyst. Such polymers are said to exhibit a pour point of - 30°C or less, as determined by ASTM Method No. D97. As set forth in the published application, polymers exhibiting such low pour points do not adversely affect the pour point of

30   a lubricant to which they are added.

Non-pourable ethylene polymers having a narrow molecular weight distribution, that is, an M_w/M_n less than 2.5, and an ultra-low molecular weight, as evidenced by a number average molecular weight (Mn) of no more than 11,000, have been heretofore unknown. Industry would find advantage in such polymers for use in adhesive formulations, and as wax

35   substitutes, ink modifiers, oil modifiers, viscosity modifiers, fibers, processing aids, sealants, caulks, etc.

5   Accordingly, the present invention further provides a non-pourable homogeneous
ultra-low molecular weight ethylene polymer which is characterized as having a number
average molecular weight (Mn), as determined by gel permeation chromatography, of no more
than 11,000, and a molecular weight distribution (M_w/M_n), as determined by gel permeation
chromatography, of from 1.5 to 2.5.

10   The present invention further provides a non-pourable homogeneous ultra-low
molecular weight ethylene polymer having longer lamella and a greater degree of crystalline
organization than corresponding higher molecular weight materials at an equivalent density.
In one instance, the present invention provides a non-pourable homogeneous ultra-low
molecular weight semicrystalline ethylene/α-olefin interpolymer having a density less than
15   0.900 g/cm^3 characterized as having lamella greater than 40 nanometers in length when
viewed using transmission electron microscopy.

        The present invention further provides a process for preparing the non-pourable
homogeneous ultra-low molecular weight ethylene polymers of the invention comprising:
reacting ethylene and at least one ethylenically unsaturated comonomer at a reaction
20   temperature of at least 80°C in the presence of a constrained geometry catalyst to form a non-
pourable homogeneous ultra-low molecular weight ethylene polymer which is characterized as
having a number average molecular weight (Mn) of no more than 11,000, and a molecular
weight distribution, M_w/M_n, as determined by gel permeation chromatography, of from 1.5 to
2.5.

25   These and other embodiments of the claimed invention are more fully set forth in the
Detailed Description set forth below.

        FIGURE 1(a) is a simplified representation of features of a transmission electron
micrograph of a homogeneous ethylene/1-octene copolymer having a density of from 0.86 to
0.88 g/cm^3 and an I_2 of 1 g/10 min.;

30   FIGURE 1(b) is a simplified representation of features of a transmission
electron micrograph of a homogeneous ethylene/1-octene copolymer having a density
of from 0.88 to 0.91 g/cm^3 and an I_2 of 1 g/10 min.;

        FIGURE 1(c) is a simplified representation of features of a transmission
electron micrograph of a homogeneous ethylene/1-octene copolymer having a density
35   of from 0.91 to 0.93 g/cm^3 and an I_2 of 1 g/10 min.;

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5   FIGURE 1(d) is a simplified representation of features of a transmission

electron micrograph of a homogeneous ethylene/1-octene copolymer having a density

greater than 0.95 g/cm^3 and an I_2 of 1 g/10 min.;

        FIGURE 2(a) is a transmission electron micrograph, at a magnification of

90,000 times, of an ethylene/1-octene copolymer having a density of 0.855 g/cm^3 and

10   an I_2 of 0.5 g/10 min.;

        FIGURE 2(b) is a transmission electron micrograph, at a magnification of

90,000 times, of the ultra-low molecular weight polymer of Example 1 (an ethylene/1-

octene copolymer having a density of 0.855 g/cm^3, an Mn of 4,600, and a melt

viscosity at 350°F of 350 centipoise);

15   FIGURE 3(a) is a transmission electron micrograph, at a magnification of

90,000 times, of the polymer of Comparative Example D (a substantially linear

ethylene/1-octene copolymer having a density of 0.870 g/cm^3 and an I_2 of 1 g/10 min);

        FIGURE 3(b) is a transmission electron micrograph, at a magnification of

90,000 times, of the polymer of Comparative Example C2 (an ethylene/1-octene

20   copolymer having a density of 0.875 g/cm^3 and an I_2 of 246 g/10 min.);

        FIGURE 3(c) is a transmission electron micrograph, at a magnification of

90,000 times, of the ultra-low molecular weight polymer of Example 2 (an ethylene/1-

octene copolymer having a density of 0.871 g/cm^3, an Mn of 9,100, and a melt

viscosity at 350°F of 4200 centipoise);

25   FIGURE 3(d) is a transmission electron micrograph, at a magnification of

90.000 times, of the ultra-low molecular weight polymer of Example 3 (an ethylene/1-

octene copolymer having a density of 0.870 g/cm^3, an Mn of 4,200, and a melt

viscosity at 350°F of 355 centipoise);

        FIGURE 4 is a transmission electron micrograph, at a magnification of 90,000

30   times, of the ultra-low molecular weight polymer of Example 4 (an ethylene/1-octene

copolymer having a density of 0.897 g/cm^3, an Mn of 8,700, and a melt viscosity at 350

°F of 5200 centipoise);

        FIGURE 4(b) is a transmission electron micrograph, at a magnification of

90,000 times, of the ultra-low molecular weight polymer of Example 17 (an ethylene/1-

5   octene copolymer having a density of 0.890 g/cm^3, an Mn of 4500, and a melt viscosity at 350°F of 350 centipoise);

FIGURE 5 is a transmission electron micrograph, at a magnification of 90,000 times, of a substantially linear ethylene/1-octene copolymer having a density of 0.915 g/cm^3 and an I$_2$ of 1 g/10 min.;

10   FIGURE 6 is a transmission electron micrograph, at a magnification of 90,000 times, of the ultra-low molecular weight polymer of Example 5 (an ethylene/1-octene copolymer having a density of 0.929 g/cm^3, an Mn of 8,900, and a melt viscosity at 350 °F of 5600 centipoise);

FIGURE 6(b)   is a transmission electron micrograph, at a magnification of 90,000 times, of the ultra-low molecular weight polymer of Example 18 (an ethylene/1-octene copolymer having a density of 0.930 g/cm^3, an Mn of 4700, and a melt viscosity at 350°F of 400 centipoise);

FIGURE 7(a) is a transmission electron micrograph, at a magnification of 90,000 times, of a substantially linear ethylene homopolymer having a density of 0.960 g/cm^3 and an I$_2$ of 1 g/10 min.; ·

FIGURE 7(b) is a transmission electron micrograph, at a magnification of 90,000 times, of the ultra-low molecular weight polymer of Example 6 (an ethylene/1-octene copolymer having a density of 0.963 g/cm^3, an Mn of 8,000, and a melt viscosity at 350°F of 5200 centipoise);

25   FIGURE 7(c) is a transmission electron micrograph, at a magnification of 90,000 times, of the ultra-low molecular weight polymer of Example 7 (an ethylene/1-octene copolymer having a density of 0.968 g/cm^3, an Mn of 3,700, and a melt viscosity at 350°F of 395 centipoise);

 FIGURE 8 is a transmission electron micrograph, at a magnification of 90,000 times, of the ultra-low molecular weight polymer of Example 13 (an ethylene/1-butene copolymer having a density of 0.868 g/cm^3 and a melt viscosity at 350°F of 5290 centipoise);

FIGURE 9 is a transmission electron micrograph, at a magnification of 90,000 times, of the ultra-low molecular weight polymer of Example 14 (an ethylene/1-butene

5   copolymer having a density of 0.887 g/cm^3 and a melt viscosity at 350°F of 5000

centipoise);

FIGURE 10 is a bar chart depicting the population of lamella having lengths in
the indicated ranges for the ethylene/octene copolymers for which the transmission
electron micrographs are set forth in FIGURES 3(a), 3(b), 3(c), and 3(d), as determined
10   by digital image analysis;

FIGURE 11 is a bar chart depicting the frequency of lamella having lengths in
the indicated ranges for the ethylene/1-octene copolymers for which the transmission
electron micrographs are set forth in FIGURES 3(a), 3(b), 3(c), and 3(d), that is, the
percentage of total lamella which have lengths in the indicated ranges, as determined by
15   digital image analysis.

FIGURE 12 is a compilation of the melting curves, as determined by
differential scanning calorimetry, for the ethylene/1-octene copolymers for which the
transmission electron micrographs are set forth in FIGURES 3(a), 3(b), 3(c), and 3(d);

FIGURE 13 is a compilation of the crystallization curves, as determined by
20   differential scanning calorimetry, for the ethylene/1-octene copolymers for which the
transmission electron micrographs are set forth in FIGURES 3(a), 3(b), 3(c), and 3(d);

FIGURE 14 is a compilation of the melting curves, as determined by
differential scanning calorimetry, for the ethylene/1-octene copolymers of Comparative
Examples G and H and of Examples 8 and 10;

25   FIGURE 15 is a compilation of the crystallization curves obtained by
differential scanning calorimetry for the ethylene/1-octene copolymers of Comparative
Examples G and H and of Examples 8 and 10;

FIGURE 16 is a plot of the total percent crystallinity of ethylene/1-octene and
ethylene/1-butene copolymers of the invention versus the density of such copolymers;
30   and

FIGURE 17 is a transmission electron micrograph, at a magnification of
90,000 times, of the ultra-low molecular weight polymer of Example 19 (an ethylene/1-
octene copolymer having a density of 0.920 g/cm^3, an Mn of 9800, and a melt
viscosity at 350°F of 5620 centipoise.

5   Unless indicated otherwise, the following testing procedures are to be
employed:

Density is measured in accordance with ASTM D-792. The samples are
annealed at ambient conditions for 24 hours before the measurement is taken.

Melt index (I_2), is measured in accordance with ASTM D-1238, condition
10   190°C/2.16 kg (formally known as "Condition (E)").

Molecular weight is determined using gel permeation chromatography (GPC)
on a Waters 150°C high temperature chromatographic unit equipped with three mixed
porosity columns (Polymer Laboratories 103, 104, 105, and 106), operating at a system
temperature of 140°C. The solvent is 1,2,4-trichlorobenzene, from which 0.3 percent
15   by weight solutions of the samples are prepared for injection. The flow rate is 1.0
mL/min. and the injection size is 100 microliters.

The molecular weight determination is deduced by using narrow molecular
weight distribution polystyrene standards (from Polymer Laboratories) in conjunction
with their elution volumes. The equivalent polyethylene molecular weights are
20   determined by using appropriate Mark-Houwink coefficients for polyethylene and
polystyrene (as described by Williams and Word in Journal of Polymer Science,
Polymer Letters, Vol. 6, (621) 1968) to derive the following equation:

$$M_{polyethylene} = a * (M_{polystyrene})^b.$$

In this equation, a = 0.4316 and b = 1.0. Weight average molecular weight, M_w, is
25   calculated in the usual manner according to the following formula: $M_w = \Sigma w_i * M_i$,
where w_i and M_i are the weight fraction and molecular weight, respectively, of the ith
fraction eluting from the GPC column.

Melt viscosity is determined in accordance with the following procedure using
a Brookfield Laboratories DVII+ Viscometer in disposable aluminum sample
30   chambers. The spindle used is a SC-31 hot-melt spindle, suitable for measuring
viscosities in the range of from 10 to 100,000 centipoise. A cutting blade is employed
to cut samples into pieces small enough to fit into the 1 inch wide, 5 inches long sample
chamber. The sample is placed in the chamber, which is in turn inserted into a
Brookfield Thermosel and locked into place with bent needle-nose pliers. The sample
35   chamber has a notch on the bottom that fits the bottom of the Brookfield Thermosel to
ensure that the chamber is not allowed to turn when the spindle is inserted and spinning.

5   The sample is heated to 350°F, with additional sample being added until the melted
      sample is about 1 inch below the top of the sample chamber. The viscometer apparatus
      is lowered and the spindle submerged into the sample chamber. Lowering is continued
      until brackets on the viscometer align on the Thermosel. The viscometer is turned on,
      and set to a shear rate which leads to a torque reading in the range of 30 to 60 percent.
10   Readings are taken every minute for about 15 minutes, or until the values stabilize,
      which final reading is recorded.

      Percent crystallinity is determined by differential scanning calorimetry using a
      Perkin-Elmer DSC 7. The percent crystallinity may be calculated with the equation :

$$\text{percent } C = (A/292 \text{ J/g}) \times 100,$$

15   wherein percent C represents the percent crystallinity and A represents the heat of
      fusion of the ethylene in Joules per gram (J/g).

      The ultra-low molecular weight ethylene polymer of the invention will be a
      homopolymer or an interpolymer of ethylene with at least one ethylenically unsaturated
      monomer, conjugated or nonconjugated diene, polyene, etc. The term "interpolymer" is
20   used herein to indicate a copolymer, or a terpolymer, or the like. That is, at least one
      other comonomer is polymerized with ethylene to make the interpolymer.

      When the ultra-low molecular weight ethylene polymer is an interpolymer,
      preferred comonomers include the C_3-C_{20} α-olefins, especially propene, isobutylene,
      1-butene, 1-hexene, 4-methyl-1-pentene, and 1-octene. Other preferred monomers
25   include styrene, or alkyl-substituted styrenes, tetrafluoroethylene,
      vinylbenzocyclobutene, 1,4-hexadiene, and naphthenics (for example, cyclopentene,
      cyclohexene and cyclooctene).

      The ultra-low molecular weight ethylene polymers of the invention will be
      characterized by a number average molecular weight of less than 11,000, preferably
30   less than 10,000. Using the process of the invention, number average molecular
      weights of less than 5000 may be obtained. However, typically, the number average
      molecular weight of the polymers will be greater than 2500.

      Number average molecular weight is related to the viscosity at 350°F of the
      ultra-low molecular weight ethylene polymers. The ultra-low molecular weight
35   ethylene polymers will be characterized by a melt viscosity at 350°F of less than 8200,

5   preferably less than 6000, with melt viscosities at 350°F of less than 600 centipoise
       being easily attained.

       Further, the number average molecular weight of the ultra-low molecular
       weight ethylene polymers is related to the melt index (I_2). Note, however, that for the
       ultra-low molecular weight ethylene polymers of the invention, melt index is not
10   measured, but is calculated from viscosity correlations. The ultra-low molecular weight
       ethylene polymers will be characterized by a calculated melt index (I_2) at 190°C of
       greater than 1000, preferably of greater than 1300, with polymers having calculated
       melt indices of at least 10,000 g/10 min. being easily attained.

       The ultra-low molecular weight ethylene polymers will typically have a density
15   of from 0.850 to 0.970 g/cm^3. The density employed will be a function of the end use
       application contemplated. For instance, when the polymer is intended as a wax
       substitute, densities greater than 0.910, preferably greater than 0.920 g/cm^3 will be
       appropriate. In contrast, when the polymer is intended as the strength-imparting
       component of an adhesive, densities less than 0.900 g/cm^3, preferably less than 0.895
20   g/cm^3 will be appropriate. When the ultra-low molecular weight ethylene polymer is
       an interpolymer of ethylene and an aromatic comonomer, such as styrene, the density of
       the interpolymer will be less than 1.10 g/cm^3.

       FIGURE 1 sets forth simplified representations of the crystalline structures of
       homogeneous ethylene/1-octene copolymers and homogeneous ethylene homopolymers
25   having an I_2 of 1 g/10 min., prepared with a monocyclopentadienyltitanium single site
       catalyst. In particular, FIGURE 1(a) depicts a homogeneous ethylene/1-octene
       copolymer having a density of from 0.86 to 0.88 g/cm^3; FIGURE 1(b) depicts a
       homogeneous ethylene/1-octene copolymer having a density of from 0.88 to 0.91
       g/cm^3; FIGURE 1(c) depicts a homogeneous ethylene/1-octene copolymer having a
30   density of from 0.91 to 0.93 g/cm^3; and FIGURE 1(d) depicts a homogeneous ethylene
       homopolymer having a density greater than 0.95 g/cm^3. The depictions set forth in
       FIGURES 1(a), 1(b), 1(c), and 1(d) are representative of what has been described as
       Type I, Type II, Type III, and Type IV morphology.

       By way of background, short chain branches from the α-olefin comonomer on
35   the ethylene/α-olefin copolymer chain are too big to be incorporated in the crystalline
       structure and thus interrupt the chain folding/bundling process. When the chain length
       between comonomer insertion points is shorter than twice the minimum thickness of
       lamellar crystallites, the polymer chains by definition can no longer crystallize via a

5   chain-folding mechanism. Rather, the chain segments between comonomer insertion points may simply bundle together to form a crystalline hard segment. These bundled chains, known as fringed micelles, have different characteristics than those crystallites formed by the chain-folding process, that is, lamella.

Theoretically, the minimum thickness of a lamellar crystallite is about 40 10   angstroms. See, for example, D. R. Burfield and N. Kashiwa, Makromol. Chem., 186, 2657 (1985). Thus, the chain length between two comonomer insertion points must be at least 80 angstroms to form one fold in the lamellar crystallite. Thus, the population, distribution, and size of the comonomer along the polymer chain will dictate the chain-folding/bundling process and the resulting crystal morphology. Polymer density is an 15   inverse function of comonomer incorporation. Accordingly, lower density polymers, having more comonomer incorporated, will have fewer carbons separating adjacent comonomer insertion points. Thus, as density decreases, the population of lamella likewise decreases.

As the density of the polymer increases, and the number of comonomer 20   insertion points decreases, the length and number of the lamella grows. Further, as the density of the polymer increases, long lamella begin to form which may cause a point of entanglement between adjacent polymer molecules. Such an entangling lamellae is referred to as a "tie chain." At even higher densities, the lamella arrange themselves as spherulites, that is, the lamella appear to radiate from common nuclei. It is believed 25   that residual catalyst provide a point of origin for the polymer chain crystallizing from the polymer melt to grow.

FIGURE 1(a) depicts what may be classified as a Type I morphology. Such a morphology is characterized by the presence of bundle-like crystals, that is, fringed micelles 101. FIGURE 1(b) depicts what may be classified as a Type II morphology. 30   Such a morphology is characterized by the presence of fringed micelles 101 and lamella 102. FIGURE 1(c) depicts what may be classified as a Type III morphology. Such a morphology is characterized by a lack of fringed micelles and by the presence of thicker lamella 102, tie chains 103, and spherulites (not shown). FIGURE 1(d) depicts what may be classified as a Type IV morphology. Such a morphology is characterized 35   by a lack of fringed micelles and tie chains, and by the presence of still thicker lamella 102 and spherulites (not shown).

The ultra-low molecular weight ethylene polymers of the invention have a crystalline structure which is markedly different from that of the higher molecular

5   weight ethylene polymers depicted in FIGURES 1(a), 1(b), 1(c), and 1(d). In particular, as evidenced by the transmission electron micrographs set forth in FIGURES 3 through 9, the ultra-low molecular weight ethylene polymers of the invention have a molecular architecture which is suggestive of more highly crystalline segments than characteristic of higher molecular weight polymers of equivalent density.

10   For instance, on the basis of FIGURE 1(a), homogeneous   ethylene/1-octene copolymers having a density of 0.870 g/cm^3 and an I_2 of 1 g/10 min. would be expected to exhibit fringed micelles, but no lamella, when viewed using transmission electron microscopy. However, (as shown in FIGURE 3(c)), ultra-low molecular weight ethylene/1-octene polymers of the invention which have a density of 0.871

15   g/cm^3 and an Mn of 9100, and (as shown in FIGURE 3(d)), ultra-low molecular weight ethylene/1-octene polymers of the invention which have a density of 0.870 g/cm^3 and an Mn of 4,300, exhibit both fringed micelles and a significant number of lamella, when viewed using transmission electron microscopy.

        Further, on the basis of FIGURE 1(d), homogeneous ethylene/1-octene
20   copolymers having a density of 0.960 g/cm^3 and an I_2 of 1 g/10 min. would be expected to exhibit lamella and spherulites, when viewed using transmission electron microscopy. However, (as shown in FIGURE 7(b)), ultra-low molecular weight ethylene/1-octene polymers of the invention which have a density of 0.963 g/cm^3 and an Mn of 8,000, and (as shown in FIGURE 7(c)), ultra-low molecular weight

25   ethylene/1-octene polymers of the invention which have a density of 0.968 g/cm^3 and an Mn of 3,700, exhibit no spherulites, but rather exhibit very long lamella, which are believed to result from epitaxial crystallization. Epitaxial crystallization refers to the growth of a crystal upon an existing crystalline substrate, wherein the newly formed crystal adopts the crystalline structure of the substrate.

30   A comparison of the transmission electron micrographs of FIGURES 2(a) and 2(b), of FIGURES 3(a), 3(b), 3(c), and 3(d), and of FIGURES 7(a), 7(b), and 7(c) indicates that as the molecular weight of the polymer decreases, the number and length of the lamella increases. For instance, FIGURE 2(b) shows that an ultra-low molecular weight ethylene/1-octene copolymer of the invention having a density of 0.855 g/cm^3

35   and an Mn of 4,600 has visually identifiable lamella (in contrast to the model for polymers having a density of 0.855 g/cm^3 set forth in FIGURE 1(a). Further, while the model set forth in FIGURE 1(c) would suggest that a copolymer having a density of 0.920 g/cm^3 would be expected to have a crystalline structure characterized by the

5   presence of lamella and spherulites, FIGURE 6 shows that an ultra-low molecular
       weight ethylene/1-octene polymer of the invention having a density of 0.929 g/cm^3 and
       an Mn of 8,900, has very long lamella, which is suggestive of epitaxial crystallization.

       The length and population of lamella for transmission electron micrographs may be
       determined by digital analysis by means known in the art. Digital images of certain of the
10   transmission electron micrographs may be acquired using a Quantimet 570 digital image
       analyzer (available from Leica, Inc.), through a CCD video camera. White top hat filters may
       be applied to the optical micrographs before detection of the binaries, that is, with the lamella
       showing white against a grey background. The filters may be varied in size as required by the
       lamellar size in the micrographs. Detection thresholds may be set by visually comparing
15   resulting binaries with the original images. Minimal editing of the binaries may be done to
       correct obvious omissions or inclusions encountered in the detection process.

       In the case of the ethylene/1-octene copolymers for which the transmission electron
       micrographs are set forth in FIGURES 3(a), 3(b), 3(c), and 3(d), the average length of the
       lamella detected and the number of lamella per cubic micrometer has been calculated. In the
20   case of FIGURE 3(a), the average lamellar length is 30 nanometers, with a population of 20
       lamella per cubic micrometer. In the case of FIGURE 3(b), the average lamellar length is 54,
       with a population of 140 lamella per cubic micrometer. In the case of FIGURE 3(c), the
       average lamellar length is 59 nanometers, with a population of 240 lamella per cubic
       micrometer. In the case of FIGURE 3(d), the average lamellar length is 66 nanometers, with a
25   population of 381 lamella per cubic micrometer. These values indicate that ultra-low
       molecular weight ethylene/1-octene polymers of the invention, which have a density of about
       0.870 g/cm^3 and Mn's of 9,100 and 4,300, respectively, have over 12 times and 40 times,
       respectively, as many lamella per cubic micrometer as do comparative polymers having a
       density of 0.870 g/cm^3 and an I_2 of 1 g/10 min.

30   FIGURE 10 is a bar chart depicting the population of lamella having lengths in the
       indicated ranges for the ethylene/octene copolymers for which the transmission electron
       micrographs are set forth in FIGURES 3(a), 3(b), 3(c), and 3(d), as determined by digital
       image analysis. Table A sets forth numerically the data used to prepare FIGURE 10.

5   Table A: Data used in Preparation of FIGURE 10

Lamellar length (nanometers)	No. of lamella per cubic micron FIGURE 3(d)	No. of lamella per cubic micron FIGURE 3(c)	No. of lamella per cubic micron FIGURE 3(b)	No. of lamella per cubic micron FIGURE 3(a)
less than 40	150	40	40	20
40 - 60	340	120	54	0
60 - 80	130	30	20	0
80 - 100	100	30	10	0
100 - 120	30	10	0	0
120-140	50	0	0	0
140 - 160	10	5	0	0
160 - 180	10	0	0	0
180 - 200	10	0	5	0

As shown in FIGURE 10 and Table A, ethylene/1-octene copolymers having a density of 0.870 g/cm^3 and an I_2 of 1 g/10 min., while they have some images with an aspect ratio greater than 3 (they have 20 lamella per cubic micron having a length less
10   than 40 nanometers), they do not have any lamella greater than 40 nanometers in length. As also shown, ethylene/1-octene copolymers having a density of 0.875 g/cm^3 and an I_2 of 246 g/10 min. have twice as many lamella less than 40 nanometers in length than copolymers having an I_2 of 1 g/10 min., and exhibit lamella in the ranges of 40-60, 60-80, and 80-100 nanometers in length (with no significant number of lamella
15   having a length greater than 100 nanometers). As also shown, ethylene/1-octene copolymers having a density of 0.871 g/cm^3 and an Mn of 9,100, have 2.2 times as many lamella having a length from 40-60 nanometers and 3 times as many lamella having a length of from 80 to 100 nanometers than ethylene/octene copolymers having a density of 0.875 g/cm^3 and an I_2 of 246 g/10 min. As also shown, ethylene/1-octene

5   copolymers having a density of 0.870 g/cm^3 and an Mn of 4,300, have over 6 times as
    many lamella having a length from 40-60 nanometers, 6 times as many lamella having
    a length from 60-80 nanometers, and 9.5 times as many lamella having a length of from
    80 to 100 nanometers than ethylene/1-octene copolymers having a density of 0.875
    g/cm^3 and an I$_2$ of 246 g/10 min. Moreover, the ethylene/octene copolymers having a
10 density of 0.870 g/cm^3 and an Mn of 4,300, have significant numbers of lamella in the
    ranges of 100-120 and 120-140 nanometers.

    FIGURE 11 is a bar chart depicting the frequency of lamella having lengths in the indicated
    ranges for the ethylene/1-octene copolymers for which the transmission electron micrographs
    are set forth in FIGURES 3(a), 3(b), 3(c), and 3(d), that is, the percentage of the total number
15 of lamella which have lengths in the indicated ranges, as determined by digital image analysis.
    Table B sets forth numerically the data used to prepare FIGURE 11.

5   Table B: Data used in Preparation of FIGURE 11

Lamellar length (nanometers)	Percent of lamella having length in indicated range FIGURE 3(d)	Percent of lamella having length in indicated range FIGURE 3(c)	Percent of lamella having length in indicated range FIGURE 3(b)	Percent of lamella having length in indicated range FIGURE 3(a)
less than 40	18	20	30	100
40 - 60	41	51	40	0
60 - 80	16	10	20	0
80 - 100	12	10	8	0
100 - 120	3	4	0	0
120-140	6	0	0	0
140 - 160	1	2	0	0
160 - 180	1	0	0	0
180 - 200	1	0	4	0

More specifically, FIGURE 11 shows that for an ultra-low density ethylene/1-
octene copolymer of the invention which has a density of 0.871 g/cm^3 and an Mn of
9,100, 80 percent of the lamella have a length greater than 40 nanometers, with 50

10   percent of the lamella having a length between 40 and 60 nanometers, over 10 percent
of the lamella having a length between 60 and 80 nanometers, and over 10 percent of
the lamella having a length between 80 and 100 nanometers. Further, FIGURE 11
shows that for an ultra-low density ethylene/1-octene copolymer of the invention which
has a density of 0.870 g/cm^3 and an Mn of 4,300, over 80 percent of the lamella have a

15   length greater than 40 nanometers, with over 40 percent of the lamella having a length
of from 40 to 60 nanometers, 16 percent of the lamella having a length of from 60 to 80
nanometers, 12 percent of the lamella having a length of from 80 to 100 nanometers,
and over 10 percent of the lamella having a length greater than 100 nanometers.

5   At higher densities, the ultra-low molecular weight ethylene polymers of the invention likewise exhibit a crystalline structure which is markedly different from that of higher molecular weight comparative materials. For instance, FIGURE 5 reveals that while an ethylene/octene copolymer having a density of 0.915 g/cm^3 and an I_2 of 1 g/10 min. has lamella, some of which appear entangled, that is, a crystalline

10   organization which corresponds to the Type III structure set forth in FIGURE 1(c). In contrast, FIGURE 6 reveals that an ethylene/octene copolymer having a density of 0.929 g/cm^3 and an Mn of 8,900, is characterized by long lamella indicative of epitaxial crystallization. The contrast between ultra-low molecular weight materials and high molecular weight materials, at higher polymer densities, is especially striking in

15   FIGURES 7(a), 7(b), and 7(c).

       The greater proportion of more highly crystalline materials (and greater proportion of more highly amorphous materials) characteristic of the ultra-low molecular weight ethylene polymers of the invention are reflected in the physical properties of the polymer, such as the melting and crystallization behavior. FIGURE 12

20   is a compilation of the melting curves obtained by differential scanning calorimetry for the ethylene/1-octene copolymers for which the transmission electron micrographs are set forth in FIGURES 3(a), 3(b), 3(c), and 3(d). FIGURE 13 is a compilation of the crystallization curves obtained by differential scanning calorimetry for the ethylene/1-octene copolymers for which the transmission electron micrographs are set forth in

25   FIGURES 3(a), 3(b), 3(c), and 3(d). FIGURE 14 is a compilation of the melting curves obtained by differential scanning calorimetry for the ethylene/1-octene copolymers of Comparative Examples G and H and of Examples 8 and 10. FIGURE 15 is a compilation of the crystallization curves obtained by differential scanning calorimetry for the ethylene/1-octene copolymers of Comparative Examples G and H and of

30   Examples 8 and 10.

       As illustrated in FIGURES 12 and 14, as the molecular weight of the copolymer decreases, the melting behavior broadens and the peak melting temperature shifts to the right. As illustrated in FIGURES 13 and 15, as the molecular weight of the copolymer decreases, the crystalline melting point likewise shifts to the right.

35   FIGURES 12 through 15 support the conclusion that the lower molecular weight of the invention have greater proportions of more highly crystalline materials (and greater portions of more highly amorphous materials) than their higher molecular weight counterparts. This suggests that the ultra-low molecular weight materials of the invention will begin to crystallize at higher temperatures than corresponding higher

5   molecular weight materials having an equivalent density. This leads to utility in
applications where the polymer or formulation must solidify quickly (such as in hot
melt adhesives) or must maintain its structural integrity upon application of heat (such
as in shoe soles which are intended for consumer machine washing and drying at
elevated temperatures).

10   Likewise, the selection of comonomer affects the high temperature
performance of the ultra-low molecular weight polymers of the invention. In particular,
as the length of the comonomer chain increases, the percent crystallinity as determined
by DSC likewise increases when density and melt index are held constant. For
instance, FIGURE 16 shows that an ethylene/1-octene polymer of the invention having

15   a density of 0.883 g/cm^3 and a melt viscosity at 350°F of 5000 centipoise (Mn of
8,200) has a greater total percent crystallinity than an ethylene/1-butene copolymer of
the invention having a density of 0.887 g/cm^3 and a melt viscosity at 350°F of 5000
centipoise, for example, 28.18 versus 26.39 percent. Accordingly, when an α-olefin
comonomer is employed, such comonomer will preferably be a C_4-C_{20} α-olefin, more

20   preferably a C_5-C_{20} α-olefin, and most preferably a C_6-C_{20} α-olefin.

The ultra-low molecular weight ethylene polymers of the invention are
characterized as being non-pourable. That is, the ultra-low molecular weight ethylene
polymers of the invention are characterized as having a pour point greater than -30°C as
determined by ASTM D-97. Preferably, the pour point of the ultra-low molecular

25   weight ethylene polymers will be greater than room temperature (25°C), and more
preferably greater than 50°C.

The ultra-low molecular weight ethylene polymers of the invention may be ethylene
homopolymers or interpolymers of ethylene and at least one suitable comonomer. Preferred
comonomers include C_{3-20} α-olefins (especially ethylene, propylene, isobutylene, 1-butene, 1-

30   hexene, 3-methyl-1-pentene, 4-methyl-1-pentene, 1-hexene, and 1-octene), C_{4-40} non-
conjugated dienes, styrene, alkyl-substituted styrene, tetrafluoroethylene, naphthenics, and
mixtures thereof.

When ethylene propylene diene terpolymers (EPDM's) are prepared, the dienes are
typically non-conjugated dienes having from 6 to 15 carbon atoms. Representative examples

35   of suitable non-conjugated dienes that may be used to prepare the terpolymers include:

(a)   Straight chain acyclic dienes such as 1,4-hexadiene; 1,5-heptadiene; and 1,6-octadiene;

5   (b)   Branched chain acyclic dienes such as 5-methyl-1,4-hexadiene; 3,7-dimethyl-1,6-
               octadiene; and 3,7-dimethyl-1,7-octadiene;

       (c)   Single ring alicyclic dienes such as 4-vinylcyclohexene; 1-allyl-4-isopropylidene
               cyclohexane; 3-allylcyclopentene; 4-allylcyclohexene; and 1-isopropenyl-4-
               butenylcyclohexene;

10   (d)   Multi-ring alicyclic fused and bridged ring dienes such as dicyclopentadiene; alkenyl,
               alkylidene, cycloalkenyl, and cycloalkylidene norbornenes, such as 5-methylene-2-
               norbornene; 5-methylene-6-methyl-2-norbornene; 5-methylene-6,6-dimethyl-2-
               norbornene; 5-propenyl-2-norbornene; 5-(3-cyclopentenyl)-2-norbornene; 5-
               ethylidene-2-norbornene; 5-cyclohexylidene-2-norbornene; etc.

15   The preferred dienes are selected from the group consisting of 1,4-hexadiene;
       dicyclopentadiene; 5-ethylidene-2-norbornene; 5-methylene-2-norbornene; 7-methyl-1,6
       octadiene; 4-vinylcyclohexene; etc. One preferred conjugated diene which may be employed
       is piperylene

               Most preferred monomers are ethylene, mixtures of ethylene, propylene and
20   ethylidenenorbornene, or mixtures of ethylene and a C_{4-8} α-olefin, more especially a C_6-C_8,
       and most especially 1-octene.

               The ultra-low molecular weight ethylene polymers of the invention may be prepared
       using a constrained geometry catalyst. Constrained geometry metal complexes and methods
       for their preparation are disclosed in U.S. Application Serial No. 545,403, filed July 3, 1990
25   (EP-A-416,815); U.S. Application Serial No. 702,475, filed May 20, 1991 (EP-A-514,828); as
       well as US-A-5,470,993, 5,374,696, 5,231,106, 5,055,438, 5,057,475, 5,096,867, 5,064,802,
       and 5,132,380. In U.S. Serial Number 720,041, filed June 24, 1991, (EP-A-514,828) certain
       borane derivatives of the foregoing constrained geometry catalysts are disclosed and a method
       for their preparation taught and claimed. In US-A 5,453,410 combinations of cationic
30   constrained geometry catalysts with an alumoxane were disclosed as suitable olefin
       polymerization catalysts.

               Exemplary constrained geometry metal complexes in which titanium is present in the
       +4 oxidation state include but are not limited to the following: (n-butylamido)dimethyl(η^5-
       tetramethylcyclopentadienyl)silanetitanium (IV) dimethyl; (n-butylamido)dimethyl(η^5-
35   tetramethylcyclopentadienyl)silanetitanium (IV) dibenzyl; (t-butylamido)dimethyl(η^5-
       tetramethylcyclopentadienyl)silanetitanium (IV) dimethyl; (t-butylamido)dimethyl(η^5-
       tetramethylcyclopentadienyl)silane-titanium (IV) dibenzyl; (cyclododecylamido)dimethyl(η^5-
       tetramethylcyclo-pentadienyl)silanetitanium (IV) dibenzyl; (2,4,6-trimethylanilido)dimethyl- (

5 η^5-tetramethylcyclopentadienyl)silanetitanium (IV) dibenzyl; (1-adamantyl-amido)dimethyl(
η^5-tetramethylcyclopentadienyl)silanetitanium (IV) dibenzyl; (t-butylamido)dimethyl(η^5-
tetramethylcyclopentadienyl)silanetitanium (IV) dimethyl; (t-butylamido)dimethyl(η^5-
tetramethylcyclopentadienyl)silanetitanium (IV) dibenzyl; (1-adamantylamido)dimethyl(η^5-
tetramethylcyclopentadienyl)-silanetitanium (IV) dimethyl; (n-butylamido)diisopropoxy(η^5-

10 tetramethylcyclo-pentadienyl)silanetitanium (IV) dimethyl; (n-butylamido)diisopropoxy(η^5-
tetramethylcyclopentadienyl)silanetitanium (IV) dibenzyl; (cyclododecylamido)-diisopropoxy(
η^5-tetramethylcyclopentadienyl)-silanetitanium (IV) dimethyl;
(cyclododecylamido)diisopropoxy(η^5-tetramethylcyclopentadienyl)-silanetitanium (IV)
dibenzyl; (2,4,6-trimethylanilido)diisopropoxy(η^5-tetramethylcyclopentadienyl)-

15 silanetitanium (IV) dimethyl; (2,4,6-trimethylanilido)diisopropoxy(η^5-tetramethyl-
cyclopentadienyl)silanetitanium (IV) dibenzyl; (cyclododecylamido)dimethoxy(η^5-
tetramethylcyclopentadienyl)silanetitanium (IV) dimethyl; (cyclododecylamido)-dimethoxy(η
^{5}-tetramethylcyclopentadienyl)silanetitanium (IV) dibenzyl; (1-
adamantylamido)diisopropoxy(η^5-tetramethylcyclopentadienyl)silanetitanium (IV) dimethyl;

20 (1-adamantylamido)diisopropoxy(η^5-tetramethylcyclopentadienyl)-silanetitanium (IV)
dibenzyl; (n-butylamido)dimethoxy(η^5-tetramethylcyclo-pentadienyl)silanetitanium (IV)
dimethyl; (n-butylamido)dimethoxy- (η^5-tetramethylcyclopentadienyl)silanetitanium (IV)
dibenzyl; (2,4,6-trimethylanilido)dimethoxy(η^5-tetramethylcyclopentadienyl)silanetitanium
(IV) dimethyl; (2,4,6-trimethylanilido)dimethoxy(η^5-tetramethylcyclopentadienyl)silane-

25 titanium (IV) dibenzyl; (1-adamantylamido)dimethoxy(η^5-tetramethylcyclo-
pentadienyl)silanetitanium (IV) dimethyl; (1-adamantylamido)dimethoxy(η^5-
tetramethylcyclopentadienyl)silanetitanium (IV) dibenzyl; (n-butylamido)-ethoxymethyl(η^5-
tetramethylcyclopentadienyl)silanetitanium (IV) dimethyl;   (n-butylamido)ethoxymethyl(
η^5-tetramethylcyclopentadienyl)silanetitanium (IV) dibenzyl;

30 (cyclododecylamido)ethoxymethyl(η^5-tetramethylcyclopentadienyl)-silanetitanium (IV)
dimethyl; (cyclododecylamido)ethoxymethyl(η^5-tetramethyl-cyclopentadienyl)silanetitaniun
(IV) dibenzyl; (2,4,6-trimethylanilido)ethoxymethyl-(η^5-
tetramethylcyclopentadienyl)silanetitanium (IV) dimethyl; (2,4,6-
trimethylanilido)ethoxymethyl(η^5-tetramethylcyclopentadienyl)silanetitanium (IV) dibenzyl;

35 (cyclododecylamido)dimethyl(η^5-tetramethylcyclopentadienyl)silane-titanium (IV) dimethyl;
(1-adamantylamido)-ethoxymethyl(η^5-tetramethylcyclo-pentadienyl)silanetitanium (IV)
dimethyl; and (1-adamantylamido)ethoxymethyl(η^5-tetramethylcyclo-
pentadienyl)silanetitanium (IV) dibenzyl.

Exemplary constrained geometry metal complexes in which titanium is present in the

40 +3 oxidation state include but are not limited to the following: (n-butylamido)dimethyl(η^5-

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5   tetramethylcyclopentadienyl)silanetitanium (III) 2-(N,N-dimethylamino)benzyl; (t-
butylamido)dimethyl(η^5-tetramethylcyclopentadienyl)silanetitanium (III) 2-(N,N-
dimethylamino)benzyl; (cyclododecylamido)dimethyl(η^5-
tetramethylcyclopentadienyl)silanetitanium (III) 2-(N,N-dimethylamino)benzyl; (2,4,6-
trimethylanilido)dimethyl(η^5-tetramethylcyclopentadienyl)silanetitanium (III) 2-(N,N-

10 dimethylamino)benzyl; (1-adamantylamido)dimethyl(η^5-
tetramethylcyclopentadienyl)silanetitanium (III) 2-(N,N-dimethylamino)benzyl; (t-
butylamido)dimethyl(η^5-tetramethylcyclopentadienyl)silanetitanium (III) 2-(N,N-
dimethylamino)benzyl; (n-butylamido)diisopropoxy(η^5-
tetramethylcyclopentadienyl)silanetitanium (III) 2-(N,N-dimethylamino)benzyl;

15 (cyclododecylamido)diisopropoxy(η^5-tetramethylcyclopentadienyl)-silanetitanium (III) 2-
(N,N-dimethylamino)benzyl; (2,4,6-trimethylanilido)diisopropoxy(η^5-2-
methylindenyl)silanetitanium (III) 2-(N,N-dimethylamino)benzyl; (1-
adamantylamido)diisopropoxy(η^5-tetramethylcyclopentadienyl)silanetitanium (III) 2-(N,N-
dimethylamino)benzyl; (n-butylamido)dimethoxy(η^5-

20 tetramethylcyclopentadienyl)silanetitanium (III) 2-(N,N-dimethylamino)benzyl;
(cyclododecylamido)dimethoxy(η^5-tetramethylcyclopentadienyl)silanetitanium (III) 2-(N,N-
dimethylamino)benzyl; (1-adamantylamido)dimethoxy(η^5-
tetramethylcyclopentadienyl)silanetitanium (III) 2-(N,N-dimethylamino)benzyl; (2,4,6-
trimethylanilido)dimethoxy(η^5-tetramethylcyclopentadienyl)silanetitanium (III) 2-(N,N-

25 dimethylamino)benzyl; (n-butylamido)ethoxymethyl(η^5-
tetramethylcyclopentadienyl)silanetitanium (III) 2-(N,N-dimethylamino)benzyl;
(cyclododecylamido)ethoxymethyl(η^5-tetramethylcyclopentadienyl)silanetitanium (III) 2-
(N,N-dimethylamino)benzyl; (2,4,6-trimethylanilido)ethoxymethyl(η^5-
tetramethylcyclopentadienyl)silanetitanium (III) 2-(N,N-dimethylamino)benzyl; and (1-

30 adamantylamido)ethoxymethyl(η^5-tetramethylcyclopentadienyl)silanetitanium (III) 2-(N,N-
dimethylamino)benzyl.

Exemplary constrained geometry metal complexes in which titanium is present in the
+2 oxidation state include but are not limited to the following: (n-butylamido)-dimethyl-(η^5-
tetramethylcyclopentadienyl)silanetitanium (II) 1,4-diphenyl-1,3-butadiene; (n-

35 butylamido)dimethyl(η^5-tetramethylcyclopentadienyl)silanetitanium (II) 1,3-pentadiene; (t-
butylamido)dimethyl(η^5-tetramethylcyclopentadienyl)silane-titanium (II) 1,4-diphenyl-1,3-
butadiene; (t-butylamido)dimethyl(η^5-tetramethyl-cyclopentadienyl)silanetitanium (II) 1,3-
pentadiene; (cyclododecylamido)dimethyl- (η^5-tetramethylcyclopentadienyl)silanetitanium
(II) 1,4-diphenyl-1,3-butadiene; (cyclododecylamido)dimethyl(η^5-

40 tetramethylcyclopentadienyl)silanetitanium (II) 1,3-pentadiene; (2,4,6-

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5   trimethylanilido)dimethyl(η^5-tetramethylcyclopentadienyl)-silanetitanium (II) 1,4-diphenyl-

1,3-butadiene; (2,4,6-trimethylanilido)dimethyl(η^5-

tetramethylcyclopentadienyl)silanetitanium (II) 1,3-pentadiene; (2,4,6-

trimethylanilido)dimethyl(η^5-tetramethylcyclopentadienyl)silanetitanium (IV) dimethyl; (1-

adamantylamido)dimethyl(η^5-tetramethylcyclopentadienyl)silane-titanium (II) 1,4-diphenyl-

10   1,3-butadiene; (1-adamantylamido)dimethyl(η^5-tetramethylcyclopentadienyl)silanetitanium

(II) 1,3-pentadiene; (t-butylamido)-dimethyl(η^5-tetramethylcyclopentadienyl)silanetitanium

(II) 1,4-diphenyl-1,3-butadiene; (t-butylamido)dimethyl(η^5-

tetramethylcyclopentadienyl)silanetitanium (II) 1,3-pentadiene; ; (n-butylamido)diisopropoxy(

η^5-tetramethylcyclopentadienyl)-silanetitanium (II) 1,4-diphenyl-1,3-butadiene; (n-

15   butylamido)diisopropoxy(η^5-tetramethylcyclopentadienyl)silanetitanium (II) 1,3-pentadiene..

(cyclododecylamido)-diisopropoxy(η^5-tetramethylcyclopentadienyl)silanetitanium (II) 1,4-

diphenyl-1,3-butadiene; (cyclododecylamido)diisopropoxy(η^5-tetramethylcyclopentadienyl)-

silanetitanium (II) 1,3-pentadiene; (2,4,6-trimethylanilido)diisopropoxy(η^5-2-methyl-

indenyl)silanetitanium (II) 1,4-diphenyl-1,3-butadiene; (2,4,6-trimethylanilido)-diisopropoxy(

20   η^5-tetramethylcyclopentadienyl)silanetitanium (II) 1,3-pentadiene; (1-

adamantylamido)diisopropoxy(η^5-tetramethylcyclopentadienyl)silanetitanium (II) 1,4-

diphenyl-1,3-butadiene; (1-adamantylamido)diisopropoxy(η^5-tetramethyl-

cyclopentadienyl)silanetitanium (II) 1,3-pentadiene; (n-butylamido)dimethoxy(η^5-

tetramethylcyclopentadienyl)silanetitanium (II) 1,4-diphenyl-1,3-butadiene; (n-

25   butylamido)dimethoxy(η^5-tetramethylcyclopentadienyl)silanetitanium (II) 1,3-pentadiene;

(cyclododecylamido)dimethoxy(η^5-tetramethylcyclopentadienyl)-silanetitanium (II) 1,4-

diphenyl-1,3-butadiene; (cyclododecylamido)dimethoxy(η^5-

tetramethylcyclopentadienyl)silanetitanium (II) 1,3-pentadiene; (2,4,6-

trimethylanilido)dimethoxy(η^5-tetramethylcyclopentadienyl)silanetitanium (II) 1,4-diphenyl-

30   1,3-butadiene; (2,4,6-trimethylanilido)dimethoxy(η^5-

tetramethylcyclopentadienyl)silanetitanium (II) 1,3-pentadiene; (1-adamantyl-

amido)dimethoxy(η^5-tetramethylcyclopentadienyl)silanetitanium (II) 1,4-diphenyl-1,3-

butadiene; (1-adamantylamido)dimethoxy(η^5-tetramethylcyclopentadienyl)-silanetitanium (II)

1,3-pentadiene; (n-butylamido)ethoxymethyl(η^5-tetramethylcyclopentadienyl)silanetitanium

35   (II) 1,4-diphenyl-1,3-butadiene; (n-butylamido)ethoxymethyl(η^5-

tetramethylcyclopentadienyl)silanetitanium (II) 1,3-pentadiene;

(cyclododecylamido)ethoxymethyl(η^5-tetramethylcyclopentadienyl)silanetitanium (II) 1,4-

diphenyl-1,3-butadiene; (cyclododecylamido)ethoxymethyl(η^5-

tetramethylcyclopentadienyl)silanetitanium (II) 1,3-pentadiene; (2,4,6-

40   trimethylanilido)ethoxymethyl(η^5-tetramethylcyclopentadienyl)silanetitanium (II) 1,4-

5   diphenyl-1,3-butadiene; (2,4,6-trimethylanilido)ethoxymethyl(η^5-
tetramethylcyclopentadienyl)silanetitanium (II) 1,3-pentadiene; (1-
adamantylamido)ethoxymethyl(η^5-tetramethylcyclopentadienyl)silanetitanium (II) 1,4-
diphenyl-1,3-butadiene; and (1-adamantylamido)ethoxymethyl(η^5-
tetramethylcyclopentadienyl)silanetitanium (II) 1,3-pentadiene.

10   The complexes can be prepared by use of well known synthetic techniques. The
reactions are conducted in a suitable noninterfering solvent at a temperature from -100 to 300
°C, preferably from -78 to 100 °C, most preferably from 0 to 50 °C. A reducing agent may be
used to cause the metal to be reduced from a higher to a lower oxidation state. Examples of
suitable reducing agents are alkali metals, alkaline earth metals, aluminum and zinc, alloys of

15   alkali metals or alkaline earth metals such as sodium/mercury amalgam and sodium/potassium
alloy, sodium naphthalenide, potassium graphite, lithium alkyls, lithium or potassium
alkadienyls, and Grignard reagents.

Suitable reaction media for the formation of the complexes include aliphatic and
aromatic hydrocarbons, ethers, and cyclic ethers, particularly branched-chain hydrocarbons

20   such as isobutane, butane, pentane, hexane, heptane, octane, and mixtures thereof; cyclic and
alicyclic hydrocarbons such as cyclohexane, cycloheptane, methylcyclohexane,
methylcycloheptane, and mixtures thereof; aromatic and hydrocarbyl-substituted aromatic
compounds such as benzene, toluene, and xylene, C_{1-4} dialkyl ethers, C_{1-4} dialkyl ether
derivatives of (poly)alkylene glycols, and tetrahydrofuran. Mixtures of the foregoing are also

25   suitable.

Suitable activating cocatalysts and activating techniques have been previously taught
with respect to different metal complexes in the following references: EP-A-277,003, US-A-
5,153,157, US-A-5,064,802, EP-A-468,651 (equivalent to U. S. Serial No. 07/547,718), EP-A-
520,732 (equivalent to U. S. Serial No. 07/876,268), WO 95/00683 (equivalent to U.S. Serial

30   No. 08/82,201), and EP-A-520,732 (equivalent to U. S. Serial No. 07/884,966 filed May 1,
1992).

Suitable activating cocatalysts for use herein include perfluorinated tri(aryl)boron
compounds, and most especially tris(pentafluorophenyl)borane; nonpolymeric, compatible,
noncoordinating, ion forming compounds (including the use of such compounds under

35   oxidizing conditions), especially the use of ammonium-, phosphonium-, oxonium-,
carbonium-, silylium- or sulfonium- salts of compatible, noncoordinating anions, and
ferrocenium salts of compatible, noncoordinating anions. Suitable activating techniques
include the use of bulk electrolysis (explained in more detail hereinafter). A combination of
the foregoing activating cocatalysts and techniques may be employed as well.

5   Illustrative, but not limiting, examples of boron compounds which may be used as an
activating cocatalysts are: tri-substituted ammonium salts such as: trimethylammonium
tetrakis(pentafluoro-phenyl) borate; triethylammonium tetrakis(pentafluorophenyl) borate;
tripropylammonium tetrakis(pentafluorophenyl) borate; tri(n-butyl)ammonium
tetrakis(pentafluorophenyl) borate; tri(sec-butyl)ammonium tetrakis(pentafluoro-phenyl)

10   borate; N,N-dimethylanilinium tetrakis(pentafluorophenyl) borate; N,N-dimethylanilinium n-
butyltris(pentafluorophenyl) borate; N,N-dimethylanilinium benzyltris(pentafluorophenyl)
borate; N,N-dimethylanilinium tetrakis(4-(t-butyldimethylsilyl)-2, 3, 5, 6-tetrafluorophenyl)
borate; N,N-dimethylanilinium tetrakis(4-(triisopropylsilyl)-2, 3, 5, 6-tetrafluorophenyl)
borate; N,N-dimethylanilinium pentafluorophenoxytris(pentafluorophenyl) borate; N,N-

15   diethylanilinium tetrakis(pentafluorophenyl) borate; N,N-dimethyl-2,4,6-trimethylanilinium
tetrakis(pentafluorophenyl) borate; trimethylammonium tetrakis(2,3,4,6-
tetrafluorophenyl)borate; triethylammonium tetrakis(2,3,4,6-tetrafluorophenyl) borate;
tripropylammonium tetrakis(2,3,4,6-tetrafluorophenyl) borate; tri(n-butyl)ammonium
tetrakis(2,3,4,6-tetrafluorophenyl) borate;

20   dimethyl(t-butyl)ammonium tetrakis(2,3,4,6-tetrafluorophenyl) borate;
N,N-dimethylanilinium tetrakis(2,3,4,6-tetrafluorophenyl) borate;
N,N-diethylanilinium tetrakis(2,3,4,6-tetrafluorophenyl) borate; and
N,N-dimethyl-2,4,6-trimethylanilinium tetrakis(2,3,4,6-tetrafluorophenyl) borate;

          disubstituted ammonium salts such as: di-(i-propyl)ammonium tetrakis(pentafluoro-
25   phenyl) borate; and dicyclohexylammonium tetrakis(pentafluorophenyl) borate;

          trisubstituted phosphonium salts such as: triphenylphosphonium tetrakis(pentafluoro-
phenyl) borate; tri(o-tolyl)phosphonium tetrakis(pentafluorophenyl) borate; and tri(2,6-
dimethylphenyl)phosphonium tetrakis(pentafluorophenyl) borate;

          disubstituted oxonium salts such as: diphenyloxonium tetrakis(pentafluoro-phenyl)
30   borate; di(o-tolyl)oxonium tetrakis(pentafluorophenyl) borate; and
di(2,6-dimethylphenyl)oxonium tetrakis(pentafluorophenyl) borate; and

          disubstituted sulfonium salts such as: diphenylsulfonium tetrakis(pentafluorophenyl)
borate; di(o-tolyl)sulfonium tetrakis(pentafluorophenyl) borate; and bis(2,6-
dimethylphenyl)sulfonium tetrakis(pentafluorophenyl) borate.

35   A most preferred activating cocatalyst is trispentafluorophenylborane.

          Alumoxanes, especially methylalumoxane or triisobutylaluminum modified
methylalumoxane are also suitable activators and may be used for activating the present metal
complexes.

5   The molar ratio of metal complex: activating cocatalyst employed preferably ranges from 1 : 1000 to 2 : 1, more preferably from 1 : 5 to 1.5 : 1, most preferably from 1 : 2 to 1 : 1. In the preferred case in which a metal complex is activated by trispentafluorophenylborane and triisobutylaluminum modified methylalumoxane, the titanium:boron:aluminum molar ratio is typically from 1 : 10 : 50 to 1 : 0.5 : 0.1, most typically from about 1 : 3 : 5.

10   A support, especially silica, alumina, or a polymer (especially poly(tetrafluoroethylene) or a polyolefin) may be employed, and desirably is employed when the catalysts are used in a gas phase polymerization process. The support is preferably employed in an amount to provide a weight ratio of catalyst (based on metal):support from 1:100,000 to 1:10, more preferably from 1:50,000 to 1:20, and most preferably from 1:10,000

15   to 1:30.

At all times, the individual ingredients as well as the recovered catalyst components must be protected from oxygen and moisture. Therefore, the catalyst components and catalysts must be prepared and recovered in an oxygen and moisture free atmosphere. Preferably, therefore, the reactions are performed in the presence of an dry, inert gas such as, for example,

20   nitrogen.

In general, the polymerization may be accomplished at conditions for Ziegler-Natta or Kaminsky-Sinn type polymerization reactions, that is, reactor pressures ranging from atmospheric to 3500 atmospheres (34.5 kPa). The reactor temperature should be greater than 80°C, typically from 100°C to 250°C, and preferably from 100°C to 150°C, with higher

25   reactor temperatures, that is, reactor temperatures greater than 100°C generally favoring the formation of lower molecular weight polymers.

In conjunction with the reactor temperature, the hydrogen:ethylene molar ratio influences the molecular weight of the polymer, with greater hydrogen levels leading to lower molecular weight polymers. When the desired polymer has an I_2 of 1 g/10 min, the

30   hydrogen:ethylene molar ratio will typically be 0 : 1. When the desired polymer has an I_2 of 1000 g/10 min., the hydrogen:ethylene molar ratio will typically be from 0.45 : 1 to 0.7 : 1. The upper limit of the hydrogen:ethylene molar ratio is about 2.2-2.5 : 1.

Generally the polymerization process is carried out with a differential pressure of ethylene of from 10 to 1000 psi (70 to 7000 kPa), most preferably from 40 to 60 psi (300 to

35   400 kPa). The polymerization is generally conducted at a temperature of from 80 to 250 °C, preferably from 90 to 170 °C, and most preferably from greater than 95 to 140 °C.

In most polymerization reactions the molar ratio of catalyst:polymerizable compounds employed is from 10^{-12}:1 to 10^{-1}:1, more preferably from 10^{-9}:1 to 10^{-5}:1.

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5   Solution polymerization conditions utilize a solvent for the respective components of
the reaction. Preferred solvents include mineral oils and the various hydrocarbons which are
liquid at reaction temperatures. Illustrative examples of useful solvents include alkanes such
as pentane, isopentane, hexane, heptane, octane and nonane, as well as mixtures of alkanes
including kerosene and Isopar E™, available from Exxon Chemicals Inc.; cycloalkanes such as
10   cyclopentane and cyclohexane; and aromatics such as benzene, toluene, xylenes, ethylbenzene
and diethylbenzene.

          The solvent will be present in an amount sufficient to prevent phase separation in the
reactor. As the solvent functions to absorb heat, less solvent leads to a less adiabatic reactor.
The solvent:ethylene ratio (weight basis) will typically be from 2.5 : 1 to 12 : 1, beyond
15   which point catalyst efficiency suffers. The most typical solvent:ethylene ratio (weight basis)
is in the range of from 5 : 1 to 10 : 1.

          The polymerization may be carried out as a batchwise or a continuous polymerization
process, with continuous polymerizations processes being required for the preparation of
substantially linear polymers. In a continuous process, ethylene, comonomer, and optionally
20   solvent and diene are continuously supplied to the reaction zone and polymer product
continuously removed therefrom.

          The ultra-low molecular weight polymers of the invention may further be made in a
slurry polymerization process, using the catalysts as described above as supported in an inert
support, such as silica. As a practical limitation, slurry polymerization takes place in liquid
25   diluents in which the polymer product is substantially insoluble. Typically, the diluent for
slurry polymerization is one or more hydrocarbons with less than 5 carbon atoms. If desired
saturated hydrocarbons such as ethane, propane or butane may be used in whole or part as the
diluent. Likewise the comonomer or a mixture of different comonomers may be used in whole
or part as the diluent. Typically, the diluent comprises in at least major part the comonomer(s)
30   to be polymerized.

          The ultra-low molecular weight polymers of the invention may be polymerized in a
first reactor, with a second polymer (of higher molecular weight and/or of different density,
and/or which is heterogeneous) being polymerized in a second reactor which is connected in
series or in parallel to that in which the ultra-low molecular weight polymer is produced, to
35   prepare in-reactor polymer blends having desirable properties. An example of a dual reactor
process which may be adapted in accordance with the teachings of this disclosure to prepare
blends wherein at least one component is an ultra-low molecular weight polymer of this
invention, is disclosed in WO 94/00500, equivalent to U. S. Serial Number 07/904,770, as well
as USSN 08/10958, filed January 29, 1993.

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5   Additives such as antioxidants (for example, hindered phenolics (for example,
Irganox™ 1010, Irganox™ 1076), phosphites (for example, Irgafos™ 168)), antiblock
additives, pigments, and fillers can also be included in the modified formulations, to the extent
that they do not interfere with the desired formulation properties.

       The skilled artisan will appreciate that the invention disclosed herein may be practiced
10   in the absence of any component which has not been specifically disclosed. The following
examples are provided as further illustration of the invention and are not to be construed as
limiting. Unless stated to the contrary all parts and percentages are expressed on a weight
basis.

<div align="center">Catalyst Preparation One</div>

15   Part 1: Preparation of TiCl3(DME)1.5

       The apparatus (referred to as R-1) was set-up in the hood and purged with nitrogen; it
consisted of a 10 L glass kettle with flush mounted bottom valve, 5-neck head,
polytetrafluoroethylene gasket, clamp, and stirrer components (bearing, shaft, and paddle).
The necks were equipped as follows: stirrer components were put on the center neck, and the
20   outer necks had a reflux condenser topped with gas inlet/outlet, an inlet for solvent, a
thermocouple, and a stopper. Dry, deoxygenated dimethoxyethane (DME) was added to the
flask (approx. 5 L). In the drybox, 700 g of TiCl3 was weighed into an equalizing powder
addition funnel; the funnel was capped, removed from the drybox, and put on the reaction
kettle in place of the stopper. The TiCl3 was added over about 10 minutes with stirring. After
25   the addition was completed, additional DME was used to wash the rest of the TiCl3 into the
flask. The addition funnel was replaced with a stopper, and the mixture heated to reflux. The
color changed from purple to pale blue. The mixture was heated for about 5 hours, cooled to
room temperature, the solid was allowed to settle, and the supernatant was decanted from the
solid. The TiCl3(DME)1.5 was left in R-1 as a pale blue solid.

30   Part 2: Preparation of [(Me4C5)SiMe2N-t-Bu][MgCl]2

       The apparatus (referred to as R-2) was set-up as described for R-1, except that flask
size was 30 L. The head was equipped with seven necks; stirrer in the center neck, and the
outer necks containing condenser topped with nitrogen inlet/outlet, vacuum adapter, reagent
addition tube, thermocouple, and stoppers. The flask was loaded with 4.5 L of toluene, 1.14 kg
35   of (Me4C5H)SiMe2NH-t-Bu, and 3.46 kg of 2 M i-PrMgCl in Et2O. The mixture was then
heated, and the ether allowed to boil off into a trap cooled to -78 °C. After four hours, the
temperature of the mixture had reached 75 °C. At the end of this time, the heater was turned

5   off and DME was added to the hot stirring solution, resulting in the formation of a white solid.
The solution was allowed to cool to room temperature, the material was allowed to settle, and
the supernatant was decanted from the solid. The $[(Me_4C_5)SiMe_2N$-t-Bu][MgCl]_2$ was left in
R-2 as an off-white solid.

Part 3: Preparation of $[(\eta^5$-Me_4C_5)SiMe_2N$-t-Bu]TiMe_2$

10   The materials in R-1 and R-2 were slurried in DME (3 L of DME in R-1 and 5 L in R-
2). The contents of R-1 were transferred to R-2 using a transfer tube connected to the bottom
valve of the 10 L flask and one of the head openings in the 30 L flask. The remaining material
in R-1 was washed over using additional DME. The mixture darkened quickly to a deep
red/brown color, and the temperature in R-2 rose from 21 °C to 32 °C. After 20 minutes, 160
15   mL of CH_2Cl_2 was added through a dropping funnel, resulting in a color change to
green/brown. This was followed by the addition of 3.46 kg of 3 M MeMgCl in THF, which
caused a temperature increase from 22 °C to 52 °C. The mixture was stirred for 30 minutes,
then 6 L of solvent was removed under vacuum. Isopar E (6 L) was added to the flask. This
vacuum/solvent addition cycle was repeated, with 4 L of solvent removed and 5 L of Isopar E
20   added. In the final vacuum step, an additional 1.2 L of solvent was removed. The material
was allowed to settle overnight, then the liquid layer decanted into another 30 L glass kettle
(R-3). The solvent in R-3 was removed under vacuum to leave a brown solid, which was re-
extracted with Isopar E; this material was transferred into a storage cylinder. Analysis
indicated that the solution (17.23 L) was 0.1534 M in titanium; this is equal to 2.644 moles of
25     $[(\eta^5$-Me_4C_5)SiMe_2N$-t-Bu]TiMe_2$. The remaining solids in R-2 were further extracted with
Isopar E, the solution was transferred to R-3, then dried under vacuum and re-extracted with
Isopar E. This solution was transferred to storage bottles; analysis indicated a concentration of
0.1403 M titanium and a volume of 4.3 L (0.6032 moles $[(\eta^5$-Me_4C_5)SiMe_2N$-t-Bu]TiMe_2$).
This gives an overall yield of 3.2469 moles of $[(\eta^5$-Me_4C_5)SiMe_2N$-t-Bu]TiMe_2$, or 1063 g.
30   This is a 72 percent yield overall based on the titanium added as $TiCl_3$.

Catalyst Preparation Two

Part 1: Preparation of $TiCl_3(DME)_{1.5}$

       The apparatus (referred to as R-1) was set-up in the hood and purged with nitrogen; t
35   consisted of a 10 L glass kettle with flush mounted bottom valve, 5-neck head,
polytetrafluoroethylene gasket, clamp, and stirrer components (bearing, shaft, and paddle).
The necks were equipped as follows: stirrer components were put on the center neck, and the
outer necks had a reflux condenser topped with gas inlet/outlet, an inlet for solvent, a

5   thermocouple, and a stopper. Dry, deoxygenated dimethoxyethane (DME) was added to the
     flask (approx. 5.2 L). In the drybox, 300 g of TiCl3 was weighed into an equalizing powder
     addition funnel; the funnel was capped, removed from the drybox, and put on the reaction
     kettle in place of the stopper. The TiCl3 was added over about 10 minutes with stirring. After
     the addition was completed, additional DME was used to wash the rest of the TiCl3 into the
10   flask. This process was then repeated with 325 g of additional TiCl3, giving a total of 625 g.
     The addition funnel was replaced with a stopper, and the mixture heated to reflux. The color
     changed from purple to pale blue. The mixture was heated for about 5 hours, cooled to room
     temperature, the solid was allowed to settle, and the supernatant was decanted from the solid.
     The TiCl3(DME)$_{1.5}$ was left in R-1 as a pale blue solid.

15   Part 2: Preparation of [(Me4C5)SiMe2N-t-Bu][MgCl]2

             The apparatus (referred to as R-2) was set-up as described for R-1, except that flask
     size was 30 L. The head was equipped with seven necks; stirrer in the center neck, and the
     outer necks containing condenser topped with nitrogen inlet/outlet, vacuum adapter, reagent
     addition tube, thermocouple, and stoppers. The flask was loaded with 7 L of toluene, 3.09 kg
20   of 2.17 M i-PrMgCl in Et2O, 250 mL of THF, and 1.03 kg of (Me4C5H)SiMe2NH-t-Bu. The
     mixture was then heated, and the ether allowed to boil off into a trap cooled to -78 °C. After
     three hours, the temperature of the mixture had reached 80 °C, at which time a white
     precipitate formed. The temperature was then increased to 90 °C over 30 minutes and held at
     this temperature for 2 hours. At the end of this time, the heater was turned off, and 2 L of
25   DME was added to the hot, stirring solution, resulting in the formation of additional
     precipitate. The solution was allowed to cool to room temperature, the material was allowed to
     settle, and the supernatant was decanted from the solid. An additional wash was done by
     adding toluene, stirring for several minutes, allowing the solids to settle, and decanting the
     toluene solution. The [(Me4C5)SiMe2N-t-Bu][MgCl]2 was left in R-2 as an off-white solid.

30   Part 3: Preparation of [(η5-Me4C5)SiMe2N-t-Bu]Ti(η4-1,3-pentadiene)

             The materials in R-1 and R-2 were slurried in DME (the total volumes of the mixtures
     were approx. 5 L in R-1 and 12 L in R-2). The contents of R-1 were transferred to R-2 using a
     transfer tube connected to the bottom valve of the 10 L flask and one of the head openings in
     the 30 L flask. The remaining material in R-1 was washed over using additional DME. The
35   mixture darkened quickly to a deep red/brown color. After 15 minutes, 1050 mL of 1,3-
     pentadiene and 2.60 kg of 2.03 M n-BuMgCl in THF were added simultaneously. The
     maximum temperature reached in the flask during this addition was 53 °C. The mixture was
     stirred for 2 hours, then approx. 11 L of solvent was removed under vacuum. Hexane was then

5   added to the flask to a total volume of 22 L. The material was allowed to settle, and the liquid

layer (12 L) was decanted into another 30 L glass kettle (R-3). An additional 15 liters of

product solution was collected by adding hexane to R-2, stirring for 50 minutes, again

allowing to settle, and decanting. This material was combined with the first extract in R-3.

The solvent in R-3 was removed under vacuum to leave a red/black solid, which was then

10 extracted with toluene. This material was transferred into a storage cylinder. Analysis

indicated that the solution (11.75 L) was 0.255 M in titanium; this is equal to 3.0 moles of $[(\eta$

5-Me4C5)SiMe2N-t-Bu]Ti(η4-1,3-pentadiene) or 1095 g. This is a 74 percent yield based cn

the titanium added as TiCl3.

Examples 1-14 and Comparative Examples C1-C4

15   The polymer products of Examples 1-14 and Comparative Examples C1-C4 are

produced in a solution polymerization process using a continuously stirred reactor. Additives

(for example, antioxidants, pigments, etc.) can be incorporated into the interpolymer products

either during the pelletization step or after manufacture, with a subsequent re-extrusion.

Examples 1-7 and Comparative Examples C1-C2 were each stabilized with 1250 ppm calcium

20 stearate, 500 ppm Irganox™ 1076 hindered polyphenol stabilizer (available from Ciba-Geigy

Corporation), and 800 ppm PEPQ (tetrakis(2,4-di-t-butylphenyl)-4,4'-biphenylene

diphosphonite) (available from Clariant Corporation). Examples 8-14 and Comparative

Examples C3-C4 were each stabilized with 500 ppm Irganox™ 1076, 800 ppm PEPQ, and 100

ppm water (as a catalyst kill agent).

25   The ethylene and the hydrogen were combined into one stream before being

introduced into the diluent mixture, a mixture of C8-C10 saturated hydrocarbons, for example,

Isopar-E hydrocarbon mixture (available from Exxon Chemical Company) and the

comonomer. In Examples 1-11 and Comparative Examples C1-C4 the comonomer was 1-

octene; in Examples 13-14, the comonomer was 1-butene; and Example 12 had no

30 comonomer. The reactor feed mixture was continuously injected into the reactor.

The metal complex and cocatalysts were combined into a single stream and were also

continuously injected into the reactor. For Examples 1-7 and Comparative Examples C1-C2,

the catalyst was as prepared in Catalyst Description One set forth above. For Examples 8-14

and Comparative Examples C2-C4, the catalyst was as prepared in Catalyst Description Two

35 set forth above. For Examples 1-14 and Comparative Examples C1-C4, the co-catalyst was

tris(pentafluorophenyl)borane, available as a 3 wt% solution in Isopar™-E mixed hydrocarbon,

from Boulder Scientific. Aluminum was provided in the form of a solution of modified

5   methylalumoxane (MMAO Type 3A) in heptane, which is available at a 2 wt percent
       aluminum concentration from Akzo Nobel Chemical Inc.

       Sufficient residence time was allowed for the metal complex and cocatalyst to react
       prior to introduction into the polymerization reactor. For the polymerization reactions of
       Examples 1-14 and Comparative Examples C1-C4, the reactor pressure was held constant at
10   about 475 psig (3380kPa). Ethylene content of the reactor, in each of Examples 1-14 and
       Comparative Examples C1-C4, after reaching steady state, was maintained at the conditions
       specified in Table One.

       After polymerization, the reactor exit stream is introduced into a separator where the
       molten polymer is separated from the unreacted comonomer(s), unreacted ethylene, unreacted
15   hydrogen, and diluent mixture stream. The molten polymer is subsequently strand chopped or
       pelletized, and, after being cooled in a water bath or pelletizer, the solid pellets are collected.
       Table I describes the polymerization conditions and the resultant polymer properties.

TABLE ONE

	C1	Ex. 1	C2	Ex. 2	Ex. 3	Ex. 4	Ex. 5	Ex. 6	Ex. 7	C3
Ethylene feed (lb/hr) (kg/hr)	2.0 (0.91)	2.0 (0.91)	2.0 (0.91)	2.0 (0.91)	2.0 (0.91)	2.0 (0.91)	3.0 (1.4)	3.0 (1.4)	3.0 (1.4)	3.0 (1.4)
Comonomer: olefin ratio (mole percent)	18.00	18.10	12.40	12.50	12.50	8.50	4.40	0.40	0.40	11.80
Hydrogen: ethylene ratio (mole percent)	0.00	1.22	0.26	0.48	1.26	0.66	0.68	0.72	1.60	0.34
Diluent: ethylene ratio (weight basis)	10.20	9.80	10.60	11.10	11.10	9.30	5.90	5.90	5.90	9.99
Catalyst metal concentration (ppm)	4	4	4	4	4	2	5	5	5	3
Catalyst flow rate (lb/hr) (kg/hr)	0.280 (0.127)	0.313 (0.142)	0.272 (0.123)	0.316 (0.143)	0.428 (0.194)	0.386 (0.175)	0.417 (0.189)	0.441 (0.200)	0.626 (0.284)	0.449 (0.203)
Co-catalyst concentration (ppm)	88	88	88	88	88	44	353	353	353	88
Co-catalyst flow rate (lb/hr) (kg/hr)	0.408 (0.185)	0.455 (0.206)	0.396 (0.180)	0.460 (0.209)	0.624 (0.283)	0.561 (0.254)	0.190 (0.086)	0.200 (0.091)	0.284 (0.129)	0.490 (0.222)
Aluminum concentration (ppm)	10	10	10	10	10	5	20	20	20	9.8
Aluminum flow rate (lb/hr) (kg/hr)	0.385 (0.174)	0.431 (0.196)	0.375 (0.170)	0.438 (0.199)	0.590 (0.268)	0.528 (0.2?0)	0.357 (0.162)	0.376 (0.171)	0.534 (0.242)	0.461 (0.209)
Reactor temperature (°C)	110	110	110	110	110	110	140	140	140	110

TABLE ONE CONTINUED

	C1	Ex. 1	C2	Ex. 2	Ex. 3	Ex. 4	Ex. 5	Ex. 6	Ex. 7	C3
Ethylene concentration in reactor exit stream (weight percent)	2.17	2.48	1.80	1.69	1.65	2.99	4.44	4.14	4.41	1.75
Polymer density (g/cm^3)	0.858	0.855	0.875	0.871	0.870	0.897	0.929	0.963	0.968	0.872
Polymer melt viscosity at 350°F (centipoise)	309000*	350	39000*	4200	355	5200	5600	5200	395	15,000
Polymer melt index (I_2 at 190°C)	32	16200*	246	1800*	16000*	1500*	1400*	1500*	14500*	583*
Polymer Mw	60,400	8,700	30,100	16,500	7,900	15,600	15,800	15,800	7,300	23,200
Polymer Mn	29,100	4,600	17,100	9,100	4,300	8,700	8,900	8,000	3,700	11,900
Polymer Mw/Mn	2.08	1.89	1.76	1.81	1.84	1.79	1.78	1.98	1.97	1.95
Peak crystallization temperature by DSC (°C)	23.73	27.13	55.73	55.44	59.05	78.57	102.76	116.01	114.76	55.73
Peak melting temperature by DSC (°C)	45.63	57	68	67	67	91.04	112.22	129.23	127.6	68
Total percent crystallinity by DSC	7.46	9.98	18.94	17.78	19.55	36.3	38.42	76.03	79.62	18.94
FIGURE No.		2(b)	3(b)	3(c)	3(d)	4		6		7(b)

TABLE ONE CONTINUED

	Ex. 8	C4	Ex. 9	Ex. 10	Ex. 11	Ex. 12	Ex. 13	Ex. 14
Ethylene feed (lb/hr) (kg/hr)	3.0 (1.4)	3.0 (1.4)	3.0 (1.4)	3.0 (1.4)	3.0 (1.4)	3.0 (1.4)	3. (1.4)	3.0 (1.4)
Comonomer : olefin ratio (mole percent)	9.10	7.40	7.40	7.30	1.24	0.00	17.10	12.70
Hydrogen : ethylene ratio (mole percent)	0.54	0.42	0.56	0.76	2.14	2.14	0.54	0.62
Diluent : ethylene ratio (weight basis)	9.99	8.59	8.59	8.59	7.69	7.70	9.99	9.00
Catalyst metal concentration (ppm)	3	3	3	3	32	32	8	8
Catalyst flow rate (lb/hr) (kg/hr)	0.450 (0.204)	0.466 (0.211)	0.555 (0.252)	0.713 (0.323)	0.304 (0.138)	0.294 (0.133)	0.392 (0.178)	0.207 (0.094)
Co-catalyst concentration (ppm)	88	88	88	88	1430	1430	353	353
Co-catalyst flow rate (lb/hr) (kg/hr)	0.490 (0.222)	0.500 (0.227)	0.605 (0.274)	0.777 (0.352)	0.219 (0.099)	0.211 (0.096)	0.278 (0.126)	0.150 (0.068)

TABLE ONE CONTINUED

	Ex. 8	C4	Ex. 9	Ex. 10	Ex. 11	Ex. 12	Ex. 13	Ex. 14
Aluminum concentration (ppm)	9.8	9.8	9.8	9.8	120.0	120.0	39.8	39.8
Aluminum flow rate (lb/hr) (kg/h)r	0.468 (0.212)	0.480 (0.218)	0.574 (0.260)	0.731 (0.332)	0.323 (0.147)	0.311 (0.141)	0.260 (0.118)	0.141 (0.064)
Reactor temperature (°C)	110	120	110	110	110	110	110	110
Ethylene concentration in reactor exit stream (weight percent)	1.71	1.41	2.17	2.48	1.80	1.69	1.65	2.99
Polymer density (g/cm³)	0.883	0.898	0.897	0.894	0.948	0.960	0.868	0.887
Polymer melt viscosity at 350°F (centipoise)	5000	15,000	5200	2500	350	512	5290	5000
Polymer melt index (I_2 at 190°C (g/10 min.))	1500*	580*	1500*	2900*	16000*	11600*		
Polymer Mw	16,200	20,300	16,100	12,000	6,900	7,400		
Polymer Mn	8,200	10,400	8,900	5,800	3,200	3,200		
Polymer Mw/Mn	1.98	1.95	1.81	2.07	2.16	2.31		
Peak crystallization temperature by DSC (°C)	69.27	79.85	78.57	81.22	109.88	116.39	47.15	65.65
Peak melting temperature by DSC (°C)	81.97	92.62	91.04	92.43	120.5	131.11	55	78.06
Total percent crystallinity by DSC	28.18	36.76	36.3	37.81	72.81	72.84	13.06	26.39
FIGURE No.							8	9

*Calculated on the basis of melt viscosity correlations in accordance with the formula:

$$I_2 = 3.6126(10 \log(\eta) - 6.6928)/-1.1363) - 9.3185,$$

where η = melt viscosity at 350°F

5   Examples 15-16 and Comparative Example C5:

The polymer products of Examples 15-16 and Comparative Example C5 were
produced in a solution polymerization process using a well-mixed recirculating loop reactor.
Each polymer was stabilized with 2000 ppm IRGANOX™ 1076 hindered polyphenol
stabilizer (available from Ciba-Geigy Corporation) and 35 ppm deionized water (as a catalyst
10   kill agent).

The ethylene and the hydrogen (as well as any ethylene and hydrogen which were
recycled from the separator, were combined into one stream before being introduced into the
diluent mixture, a mixture of C_8-C_{10} saturated hydrocarbons, for example, ISOPAR™-E
(available from Exxon Chemical Company) and the comonomer 1-octene.

15   The metal complex and cocatalysts were combined into a single stream and were also
continuously injected into the reactor. The catalyst was as prepared in Catalyst Description
Two set forth above; the primary cocatalyst was tri(pentafluorophenyl)borane, available from
Boulder Scientific as a 3 wt percent solution in ISOPAR-E mixed hydrocarbon; and the
secondary cocatalyst was modified methylalumoxane (MMAO Type 3A), available from Akzo
20   Nobel Chemical Inc. as a solution in heptane having 2 wt percent aluminum.

Sufficient residence time was allowed for the metal complex and cocatalyst to react
prior to introduction into the polymerization reactor. The reactor pressure was held constant at
about 475 psig (3380 kPa).

After polymerization, the reactor exit stream was introduced into a separator where the
25   molten polymer was separated from the unreacted comonomer(s), unreacted ethylene,
unreacted hydrogen, and diluent mixture stream, which was in turn recycled for combination
with fresh comonomer, ethylene, hydrogen, and diluent, for introduction into the reactor. The
molten polymer was subsequently strand chopped or pelletized, and, after being cooled in a
water bath or pelletizer, the solid pellets were collected. Table Two describes the
30   polymerization conditions and the resultant polymer properties.

5

Table Two

	C5	Ex. 15	Ex. 16
Ethylene fresh feed rate (lbs/hr) (kg/hr)	140 (63.5)	140 (63.5)	140 (63.5)
Total ethylene feed rate (lbs/hr) (kg/hr)	146.2 (66.32)	146.17 (66.30)	146.5 (66.45)
Fresh octene feed rate (lbs/hr) (kg/hr)	45.4 (20.6)	49.5 (22.4)	12.67 (5.75)
Total octene feed rate (lbs/hr) (kg/hr)	Not determined	112 (50.8)	32.9 (14.9)
Total octene concentration (weight percent)	Not determined	11.4	3.36
Fresh hydrogen feed rate (standard cm^3/min)	4025	5350	16100
Solvent and octene feed rate (lbs/hr) (kg/hr)	840 (381)	839.4 (380.8)	840 (381)
Ethylene conversion rate (wt percent)	90.7	90.3	88.26
Reactor temperature (°C)	109.86	119.8	134.3
Feed temperature (°C)	15	15	15.3
Catalyst concentration (ppm)	70	70	70
Catalyst flow rate (lbs/hr) (kg/hr)	0.725 (0.329)	1.265 (0.5738)	4.6 (2.1)
Primary cocatalyst concentration (ppm)	1200	2031	1998
Primary cocatalyst flow rate (lbs/hr) (kg/hr)	2.96 (1.34)	1.635 (0.7416)	5.86 (2.66)
Primary cocatalyst to catalyst molar ratio (B:Ti)	2.96	3.48	2.897
Secondary cocatalyst concentration (ppm)	198	198	198
Secondary cocatalyst flow rate (lbs/hr) (kg/hr)	0.718 (0.326)	1.258 (0.571)	3.7 (1.7)
Secondary cocatalyst to catalyst molar ratio (Al:Ti)	5	4.986	4.037
Product density (g/cm^3)	0.8926	0.8925	0.9369
Product melt viscosity at 350°F (centipoise)	12,500	4,000	400
Polymer melt index (I_2 at 190°C)*	686*	1,900*	14,000*
Polymer Mn	12,300*	8,900*	4,700*

* Calculated on the basis of melt viscosity correlations in accordance with the formulas:

$$I_2 = 3.6126(10^{\log(\eta)-6.6928)/-1.1363}) - 9.3185,$$

$$Mn = 10[(\log\eta + 10.46)/3.56)]$$

where η = melt viscosity at 350°F.

10

5

Except as noted, Examples 17-19 were prepared in accordance with the procedure set forth above with respect to Examples 1-14. In particular, Examples 17 and 18 were prepared using a catalyst prepared in accordance with Catalyst Procedure 2. The additives employed were 1000 ppm Irganox™ 1076 hindered polyphenol stabilizer (available from Ciba-Geigy

10   Corporation) and 100 ppm water. In the case of Example 18, ethylbenzene, rather than Isopar™ E mixed hydrocarbon, was utilized as the solvent.

Example 19 was prepared using a catalyst prepared in accordance with Catalyst Procedure 1. The additives employed were 1250 ppm calcium stearate, 500 ppm Irganox™ 1076 hindered polyphenol stabilizer (available from Ciba-Geigy Corporation), and 800 ppm

15   PEPQ (tetrakis(2,4-di-t-butylphenyl)-4,4'-biphenylene diphosphonite) (available from Clariant Corporation).

The run conditions employed and a description of the resultant polymers is set forth in the following Table Three:

5

TABLE THREE

	Ex. 17	Ex. 18	Ex. 19
Ethylene fresh feed rate (lbs/hr) (kg/hr)	2.5 (1.1)	3.5 (1.6)	3.02 (1.37)
Total ethylene feed rate (lbs/hr) (kg/hr)	2.5 (1.1)	3.5 (1.6)	3.02 (1.37)
Fresh octene feed rate (lbs/hr) (kg/hr)	1.9 (0.86)	1.52 (0.689)	1.1 (0.50)
Total octene feed rate (lbs/hr)	1.9	1.52	1.1
Total octene concentration (weight percent)	11.44	6.47	5.52
Fresh hydrogen feed rate (standard cm^3/min)	199.9	292.4	124.9
Solvent and octene feed rate (lbs/hr) (kg/hr)	14.1 (6.40)	20.04 (9.253)	16.9 (7.66)
Ethylene conversion rate (wt percent)	75.2	85.5	69.3
Reactor temperature (°C)	119.8	136.3	140.4
Feed temperature (°C)	26.9	33.93	40
Catalyst concentration (ppm)	12	2.4	5
Catalyst flow rate (lbs/hr) (kg/hr)	0.4543 (0.2061)	0.60717 (0.27541)	0.4174 (0.1893)
Primary cocatalyst concentration (ppm)	92	92	393
Primary cocatalyst flow rate (lbs/hr) (kg/hr)	0.67 (0.30)	0.3664 (0.1662)	0.18967 (0.08603)
Primary cocatalyst to catalyst molar ratio (B:Ti)	-	2.16	3.3
Secondary cocatalyst concentration (ppm)	-	21.74	19.78
Secondary cocatalyst flow rate (lbs/hr) (kg/hr)	-	0.302 (0.137)	0.3569 (0.1619)
Secondary cocatalyst to catalyst molar ratio (Al:Ti)		8	6
Product density (g/cm^3)	0.890	0.930	0.920
Product melt viscosity at 350°F (centipoise)	350	400	5620
Polymer melt index (I_2 at 190°C)*	16,000	14,000	1400
Polymer Mn*	4500	4700	9800

* Calculated on the basis of melt viscosity correlations in accordance with the formulas:

$$I_2 = 3.6126(10^{(\log(\eta)-6.6928)/-1.1363}) - 9.3185,$$

$$Mn = 10^{[(\log \eta + 10.46)/3.56)]}$$

10   where η melt viscosity at 350°F.

5   Comparative Examples

To a 4 liter autoclave stirred reactor, 865.9 g of ISOPAR™-E hydrocarbon (available from Exxon Chemical Company) and 800.4 g 1-octene were charged. The reactor was heated to 120°C and hydrogen was added from a 75 cc cylinder. Hydrogen was added to cause a 250 psig (1800 kPa) pressure drop in the cylinder. The reactor was then pressurized to 450 psig
10   (3200 kPa) of ethylene. Catalyst was added at the rate of 1 cc/min. The catalyst was as prepared in the Catalyst One Preparation set forth above and was mixed with other co-catalysts at a ratio of 1.5 mL of a 0.005 M of Catalyst Preparation One, 1.5 mL of a 0.015 M solution of tris(pentafluorophenyl)borane in ISOPAR-E hydrocarbon mixture (a 3 wt percent solution of tris(pentafluorophenyl)borane in ISOPAR-E hydrocarbon mixture is available from Boulder
15   Scientific), 1.5 mL of a 0.05 M solution of modified methylalumoxane in ISOPAR-E hydrocarbon mixture (MMAO Type 3A) (a solution of MMAO Type 3A in heptane with a 2 wt percent aluminum content is available from Akzo Nobel Chemical Inc.), and 19.5 mL of ISOPAR-E hydrocarbon mixture. Ethylene was supplied on demand. The reactor temperature and pressure were set at 120°C and 450 psig (3200 kPa), respectively. The reaction continued
20   for 23.1 minutes. At this time, the agitation was stopped and the reactor contents transferred to a glass collection kettle. The reactor product was dried in a vacuum oven overnight.

The ethylene/octene product thus prepared had a density of 0.867 g/cm^3, and an I_2 at 190°C of 842 g/10 min.

The following additional comparative examples represent ethylene/1-octene
25   substantially linear polymers prepared in accordance with the teachings of U.S. Patent Nos. 5,272,236 and 5,278,272. A description of the comparative examples, as well as some representative properties, is set forth in Table Four.

5

Table Four

	Density (g/cm^3)	Melt index at 190°C (g/10 min.)	Peak crystallization temperature by DSC (°C)	Peak melting temperature by DSC (°C)	Total percent crystallinity by DSC
Comparative Ex. A	0.863	0.5	32.98	50.07	12.3
Comparative Ex. B	0.863	14	39.84	57.41	13.95
Comparative Ex. C	0.868	0.5	42.73	56.3	15.65
Comparative Ex. D	0.87	1.0	47.24	55.34	13.5
Comparative Ex. E	0.87	5	45.6	63.44	17.05
Comparative Ex. F	0.87	30	49.13	60.72	18.62
Comparative Ex. G	0.885	1	62.29	80.11	26.57
Comparative Ex. H	0.885	30	66.63	84.43	28.15
Comparative Ex. I	0.902	30	82.47	98.78	40.41
Comparative Ex. J	0.902	4.3	80.84	99.04	39.14
Comparative Ex. K	0.903	1	82.97	99.49	36.23
Comparative Ex. L	0.915	1	95.78	109.0	47.91

Transmission Electron Micrograph Preparation and Digital Analysis Thereof

Transmission electron micrographs are taken of the specific polymers of the Examples and Comparative Examples, and are set forth in the FIGURES above. In each case, the

10   polymers were formed into compression molded plaques having a thickness of 125 mils (0.318 cm) and a diameter of 1 inch (2.5 cm). The plaques were cooled at the rate of 15°C/minute. The crystalline structure was revealed by preferential oxidation of the amorphous polyethylene by ruthenium tetraoxide. The polymer films were exposed for 120 minutes to ruthenium tetraoxide vapors generated from a solution of 0.2 g ruthenium chloride and 10 mL of a 5.35

15   weight percent solution of sodium hypochlorite in 100 mL of water. Sections of the plaque having a thickness of 1000 angstroms were cut at room temperature with a Reichert Jung Ultracut E microtome and placed on a 200 copper mesh grid having a polyvinyl Formvar support (the support is available from Electron Microscopy Sciences). Microscopy was done

5   on a JEOL 2000FX TEM operated at an accelerating voltage of 100 kilovolts. The resultant micrographs are set forth in the FIGURES, with 1 mm representing 0.01111 micrometer.

Digital images of certain of the transmission electron micrographs were acquired using a Quantimet 570 digital image analyzer (available from Leica, Inc.), through a CCD video camera. White top hat filters were applied to the optical micrographs before detection of the
10   binaries, that is, the lamella showed white against a grey background. The filters were disks about 6 nanometers in size. Detection thresholds were set by visually comparing resulting binaries with the original images. Minimal editing of the binaries was done to correct obvious omissions or inclusions encountered in the detection process.

The lengths of the depicted lamella were measured. The lamella in each of the
15   following ranges of length were counted: less than 40 nanometers, 40-60 nanometers, 60-80 nanometers, 80-100 nanometers, 100-120 nanometers, 120-140 nanometers, 140-160 nanometers, 160-180 nanometers, 180-200 nanometers, and greater than 200 nanometers. The average lamellar length was determined. As all lamella in the section were in focus, that is, there were no lamella hidden by other lamella, the number of lamella per cubic micron was
20   determined by multiplying the number of lamella per square micron by the section thickness, that is, 1000 angstroms.

5

1. A non-pourable homogeneous ultra-low molecular weight ethylene polymer which is characterized as having a number average molecular weight (Mn) as determined by gel permeation chromatography, of no more than 11,000, and a molecular weight distribution, M_w/M_n, as determined by gel permeation chromatography, of from 1.5 to

10   2.5.

2. The ultra-low molecular weight ethylene polymer of Claim 1, wherein said polymer is a copolymer of ethylene and at least one comonomer selected from the group consisting of ethylenically unsaturated monomers, conjugated or nonconjugated dienes, and polyenes.

15 3. The ultra-low molecular weight ethylene polymer of Claim 2, wherein said comonomer is an ethylenically unsaturated monomer selected from the group consisting of the C_3-C_{20} α-olefins, styrene, alkyl-substituted styrene, vinylbenzocyclobutane, 1,4-hexadiene, and naphthenics.

4. The ultra-low molecular weight ethylene polymer of Claim 3, wherein the comonomer

20   is an ethylenically unsaturated monomer which is a C_3-C_{20} α-olefin, and wherein the α-olefin is further selected from the group consisting of 1-propene, isobutylene, 1-butene, 1-hexene, 1-heptene, 4-methyl-1-pentene, and 1-octene.

5. The ultra-low molecular weight ethylene polymer of Claim 3, wherein the comonomer is an ethylenically unsaturated monomer which is a C_3-C_{20} α-olefin, and wherein the

25   α-olefin is further selected from the group consisting of 1-butene, 1-hexene, 4-methyl-1-pentene, 1-heptene, and 1-octene.

6. The ultra-low molecular weight ethylene polymer of Claim 3, wherein the comonomer is an ethylenically unsaturated monomer which is a C_3-C_{20} α-olefin, and wherein the α-olefin is further selected from the group consisting of 1-hexene, 4-methyl-1-pentene,

30   1-heptene, and 1-octene.

7. The ultra-low molecular weight ethylene polymer of Claim 1, wherein the polymer has a density of from 0.850 to 0.869 g/cm^3.

8. The ultra-low molecular weight ethylene polymer of Claim 1, wherein the polymer has a density of from 0.870 to 0.889 g/cm^3.

-41-

9. The ultra-low molecular weight ethylene polymer of Claim 1, wherein the polymer has a density of from 0.890 to 0.899 g/cm^3.

10. The ultra-low molecular weight ethylene polymer of Claim 1, wherein the polymer has a density of from 0.900 to 0.919 g/cm^3.

11. The ultra-low molecular weight ethylene polymer of Claim 1, wherein the polymer has a density of from 0.920 to 0.949 g/cm^3.

12. The ultra-low molecular weight ethylene polymer of Claim 1, wherein the polymer has a density greater than 0.950 g/cm^3.

13. A non-pourable homogeneous ultra-low molecular weight semicrystalline ethylene/α-olefin interpolymer having a density less than 0.900 g/cm^3 characterized as having lamella greater than 40 nanometers in length when viewed using transmission electron microscopy.

14. The ultra-low molecular weight semicrystalline ethylene/α-olefin interpolymer of Claim 13, wherein at least 60 percent of the lamella have a length greater than 40 nanometers.

15. The ultra-low molecular weight semicrystalline ethylene/α-olefin interpolymer of Claim 14, wherein at least 20 percent of the lamella have a length greater than 60 nanometers.

16. The ultra-low molecular weight semicrystalline ethylene/α-olefin interpolymer of Claim 13, wherein at least 80 percent of the lamella have a length greater than 40 nanometers.

17. The ultra-low molecular weight semicrystalline ethylene/α-olefin interpolymer of Claim 16, wherein at least 30 percent of the lamella have a length greater than 60 nanometers.

18. The ultra-low molecular weight semicrystalline ethylene/α-olefin interpolymer of Claim 16, wherein at least 10 percent of the lamella have a length greater than 80 nanometers.

19. The ultra-low molecular weight semicrystalline ethylene/α-olefin interpolymer of Claim 16, wherein at least 40 percent of the lamella have a length greater than 60 nanometers.

5   20.   The ultra-low molecular weight semicrystalline ethylene/α-olefin interpolymer of
            Claim 16, wherein at least 20 percent of the lamella have a length greater than 80
            nanometers.

     21.   The ultra-low molecular weight semicrystalline ethylene/α-olefin interpolymer of
            Claim 16, wherein at least 5 percent of the lamella have a length greater than 100
10   nanometers.

     22.   A non-pourable homogeneous ultra-low molecular weight ethylene polymer having a
            density of at least 0.920 g/cm^3, which is characterized as lacking spherulites and as
            having lamella with an average length greater than 100 nanometers when viewed using
            transmission electron microscopy.

15   23.   A process comprising reacting ethylene and at least one ethylenically unsaturated
            comonomer at a reaction temperature of at least 80°C in the presence of a constrained
            geometry catalyst to form a non-pourable homogeneous ultra-low molecular weight
            ethylene polymer which is characterized as having a number average molecular weight
            (Mn) of no more than 11,000, and a molecular weight distribution, M_w/M_n, as
20   determined by gel permeation chromatography, of from 1.5 to 2.5.

Figure 1(a)

101

101

Figure 1(b)

102

103
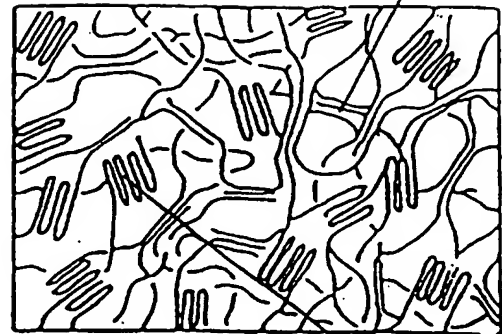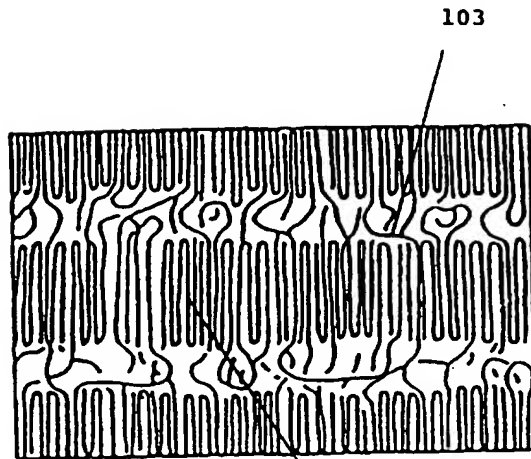
Figure 1(c)

102
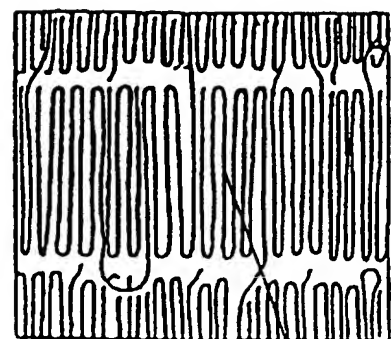
Figure 1(d)

102
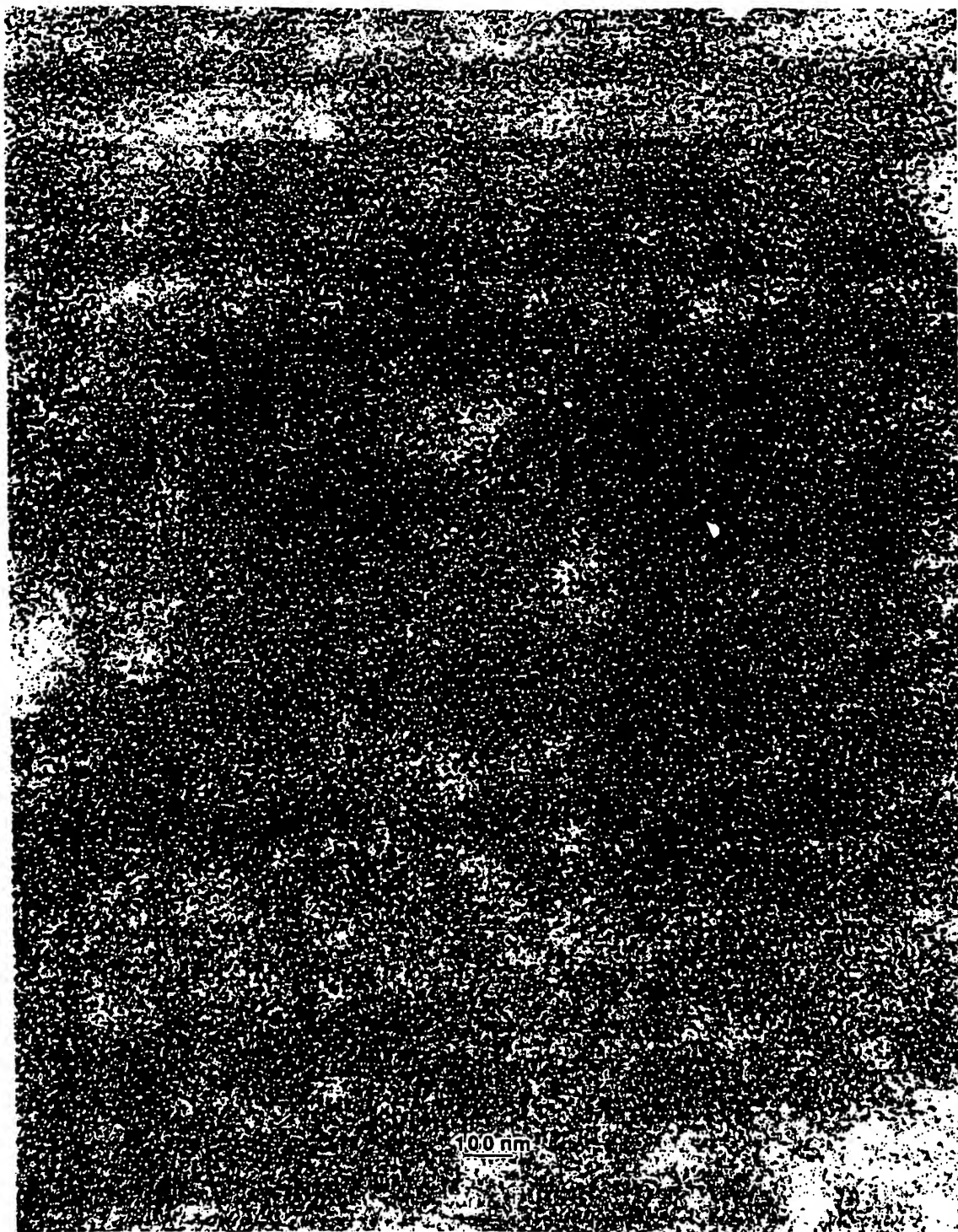
FIGURE 2(a)
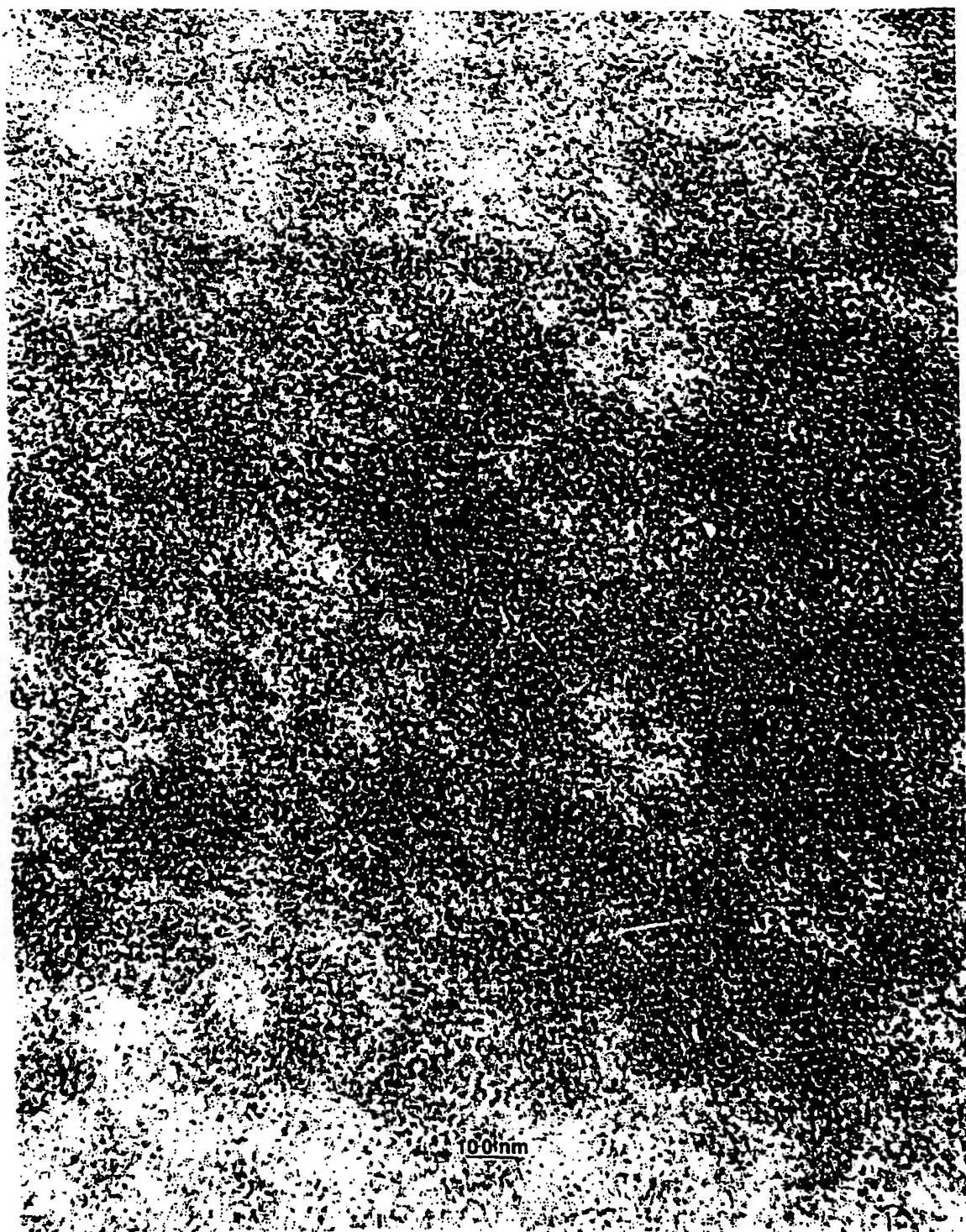
FIGURE 2(b)
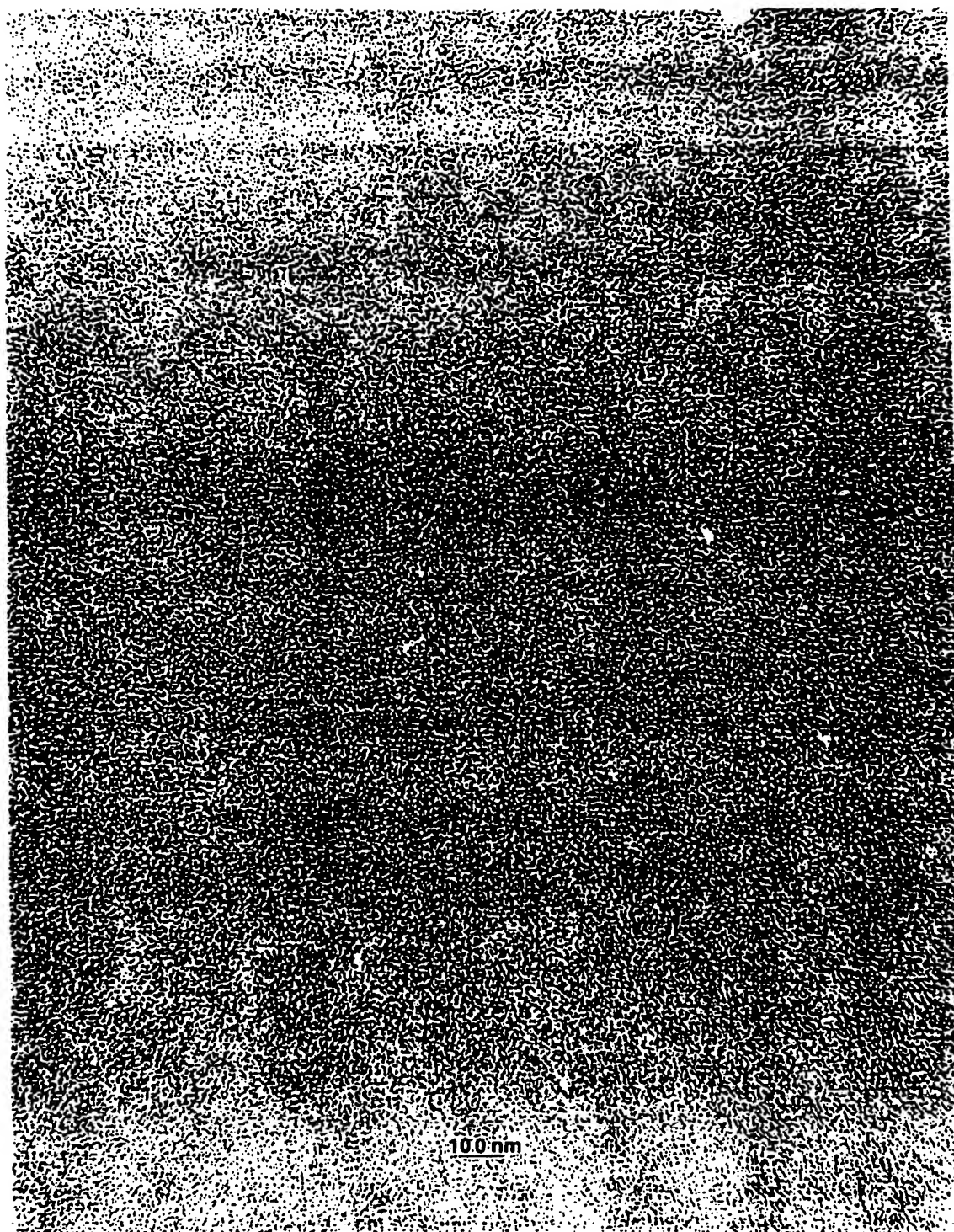
FIGURE 3(a)
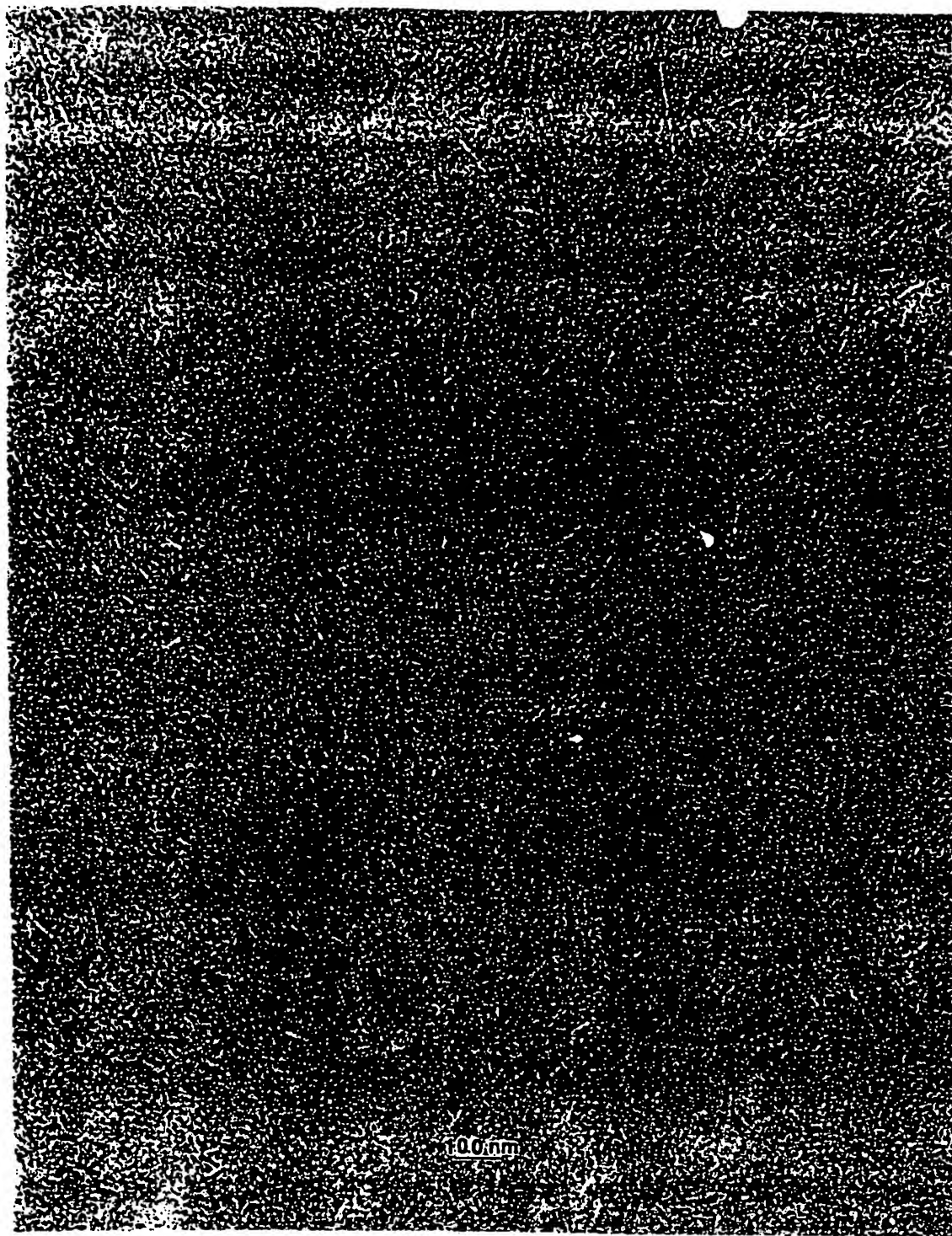
FIGURE 3(b)
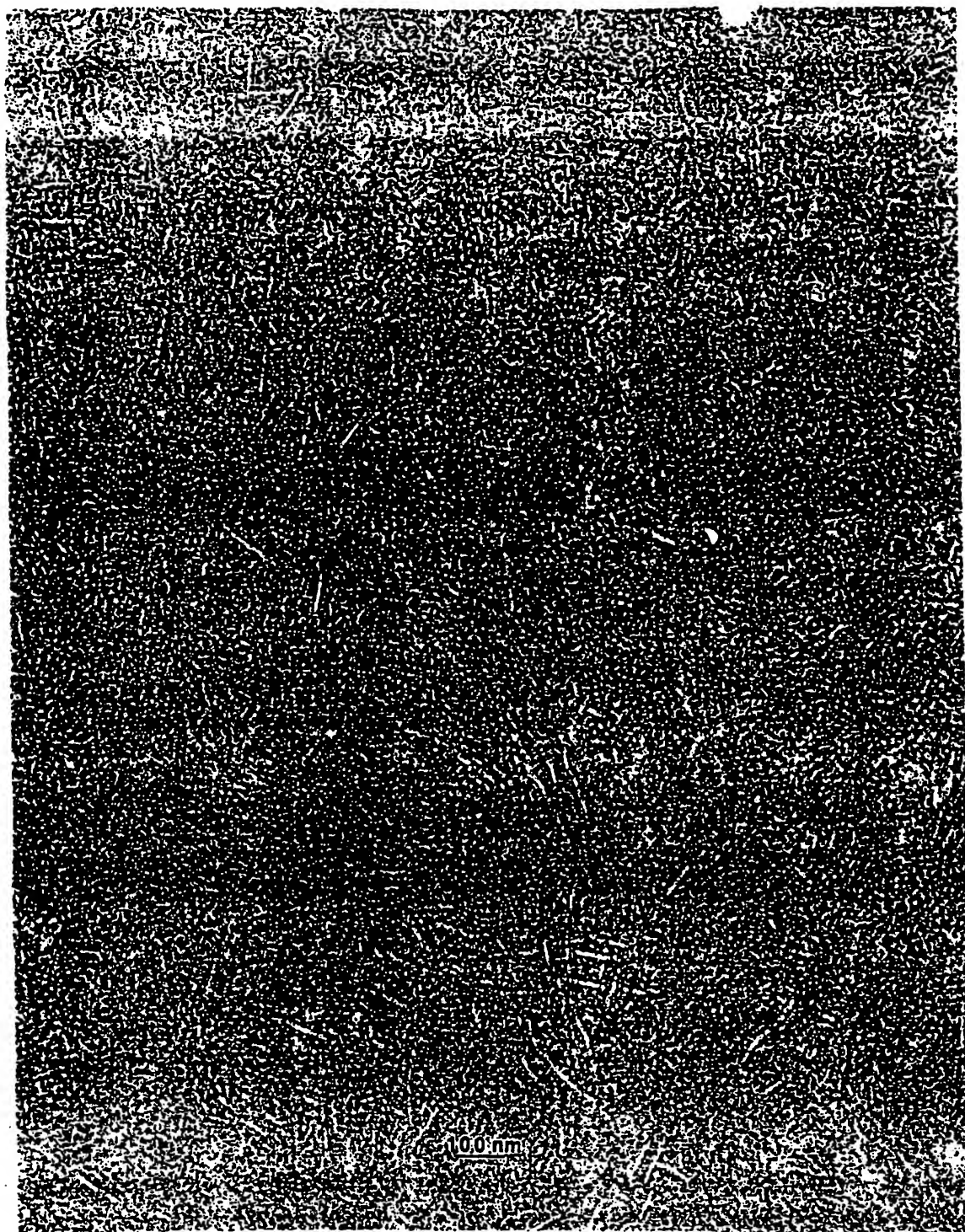
FIGURE 3(c)
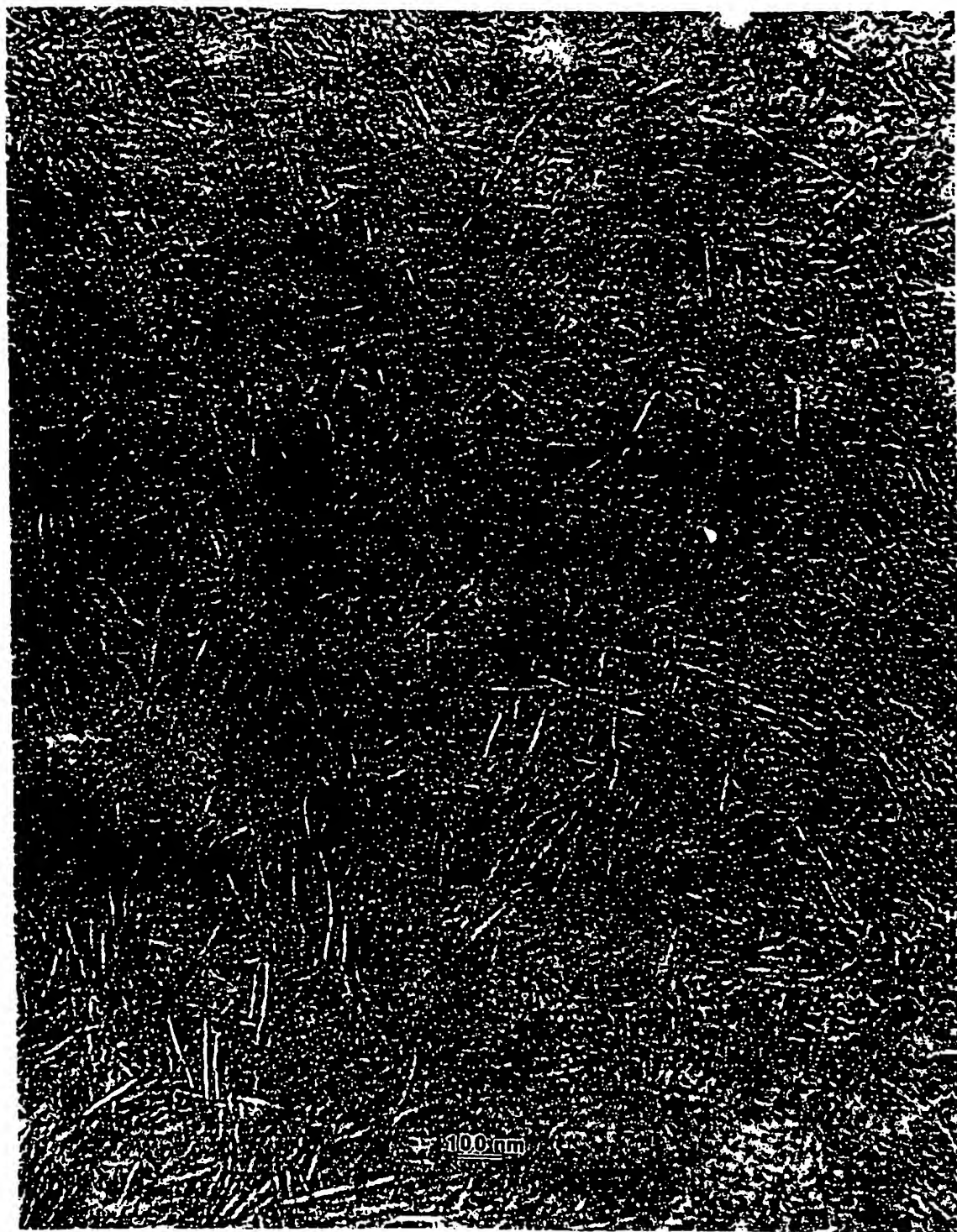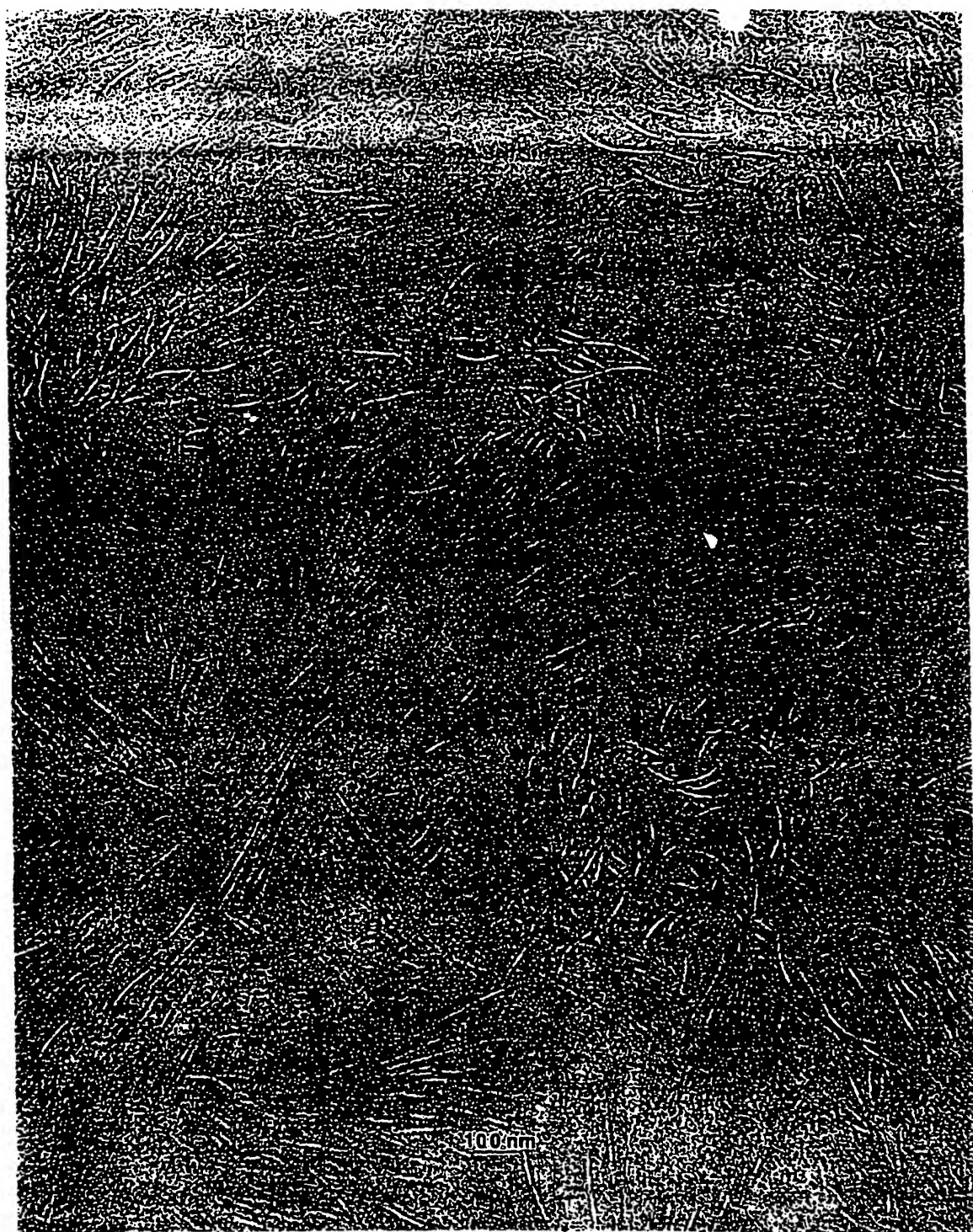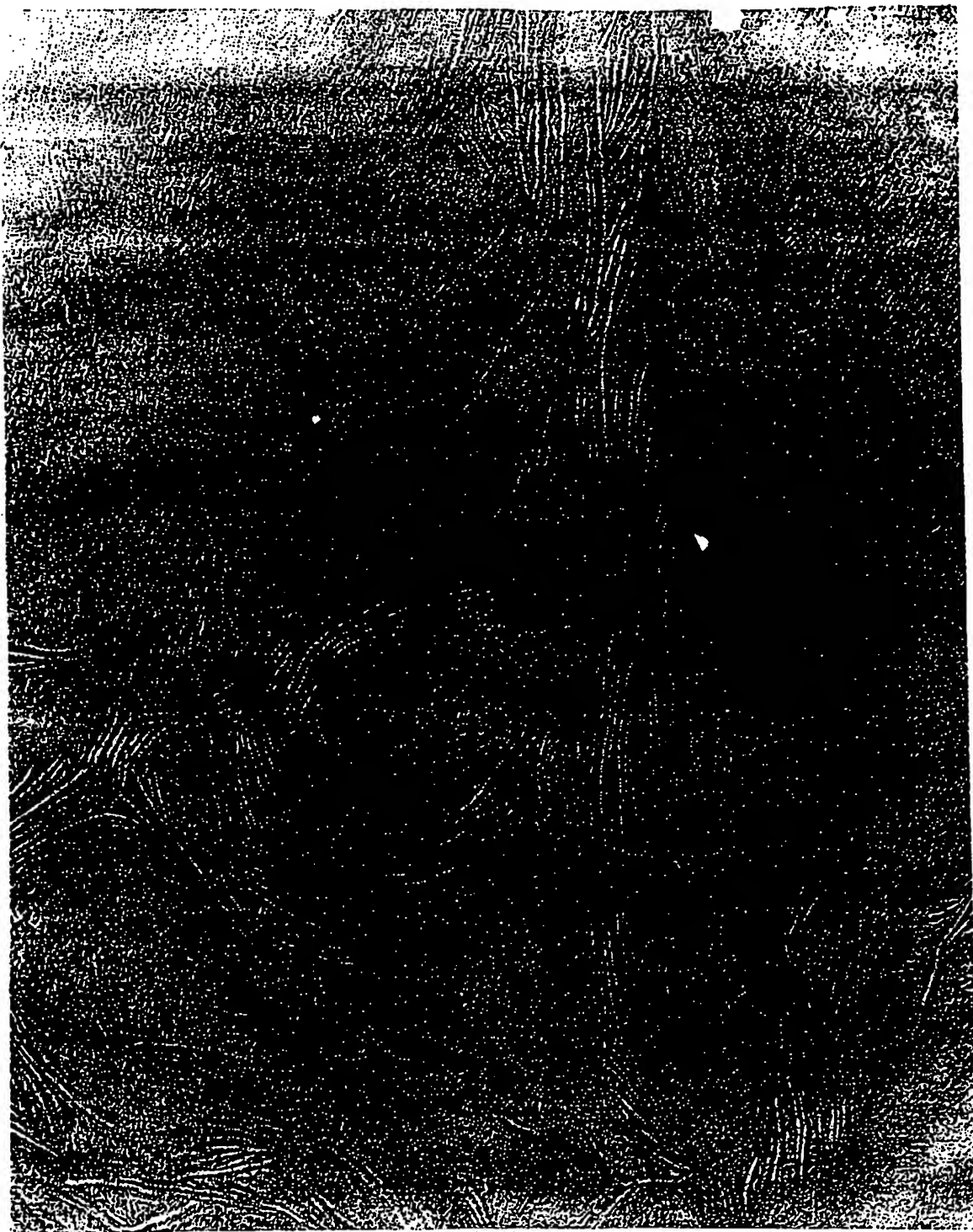
FIGURE 3(d)
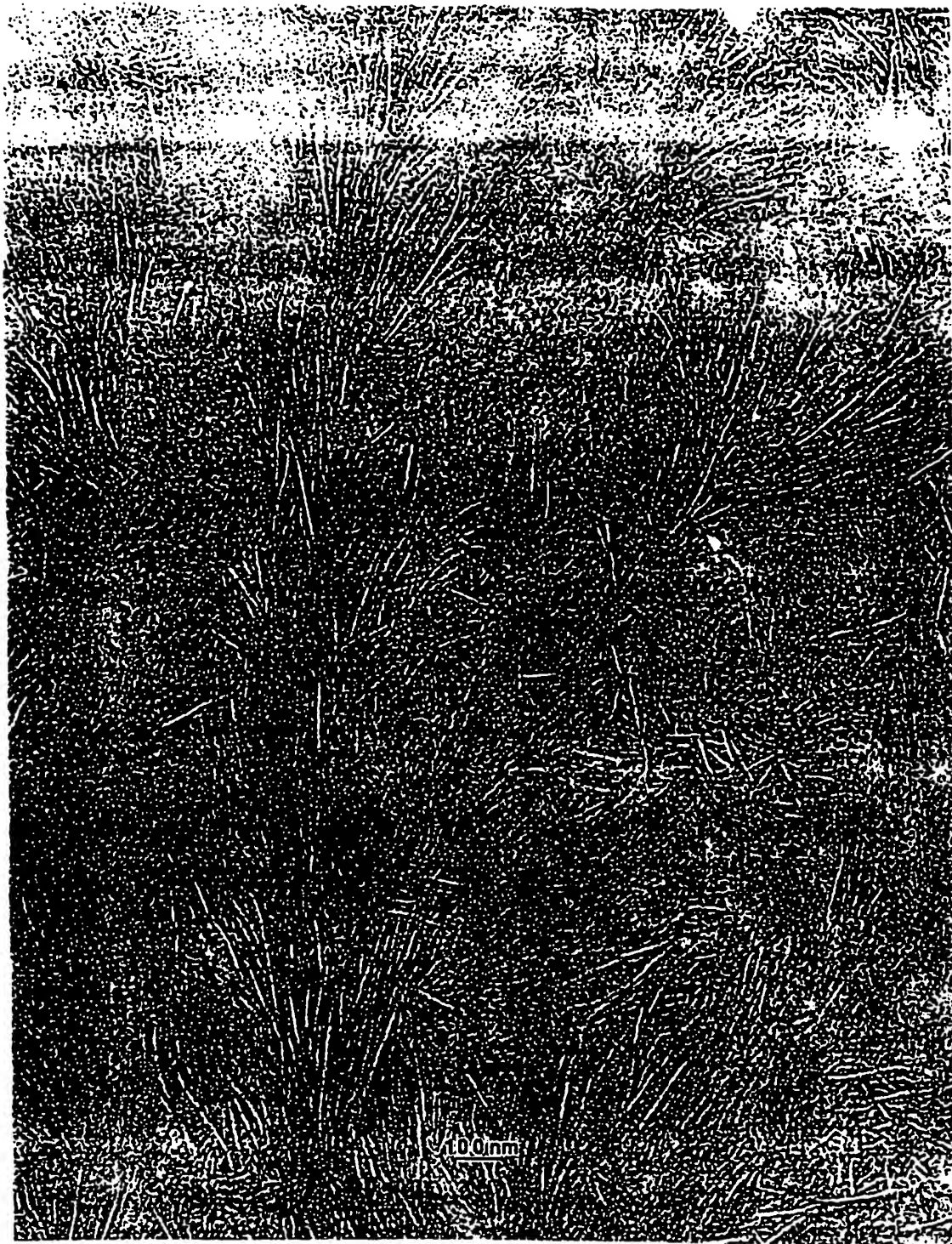
FIGURE 4

FIGURE 4(b)

FIGURE 5

FIGURE 6

FIGURE 6(b)

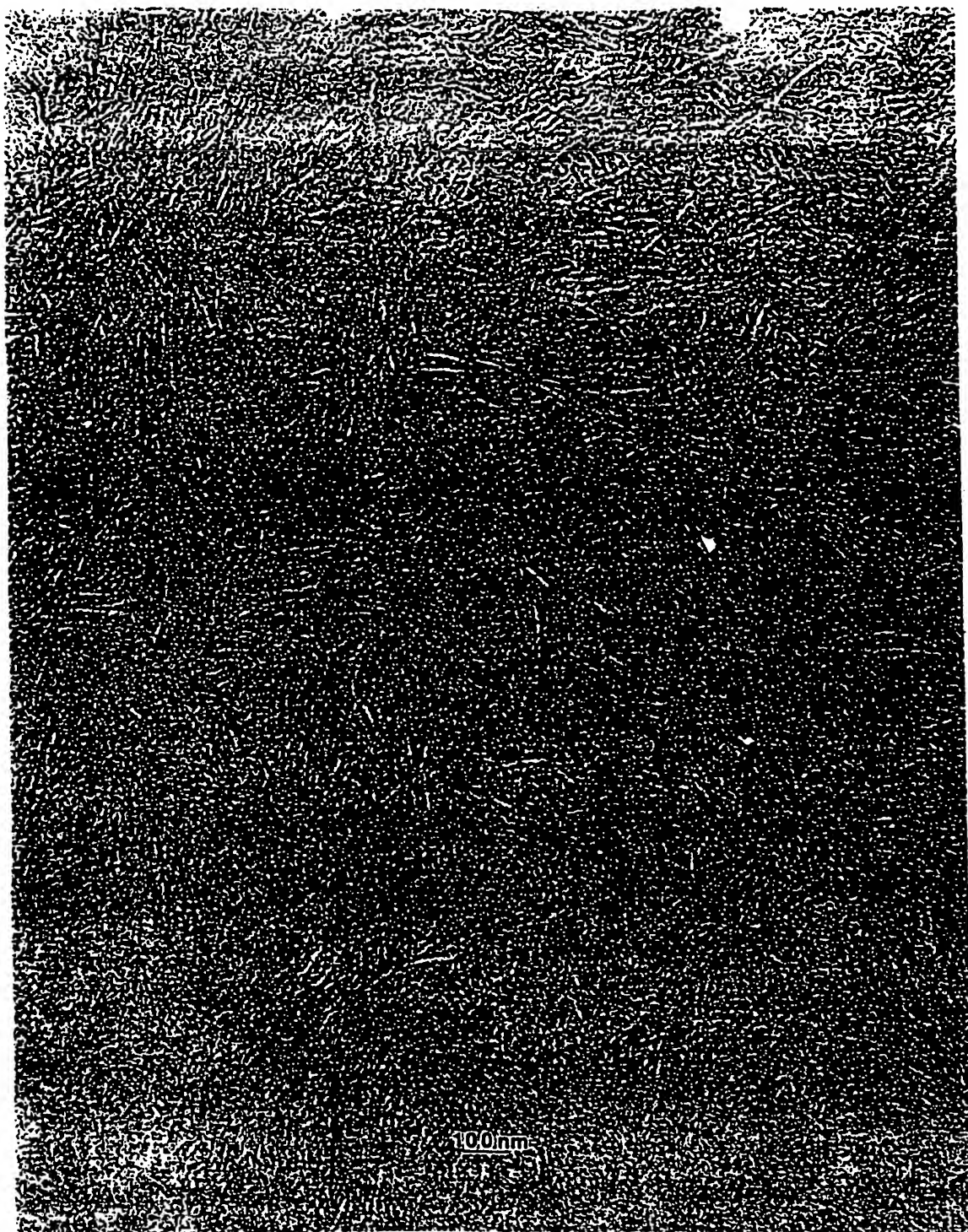FIGURE 7(a)

10.0 nm

FIGURE 7(b)

FIGURE 7(c)
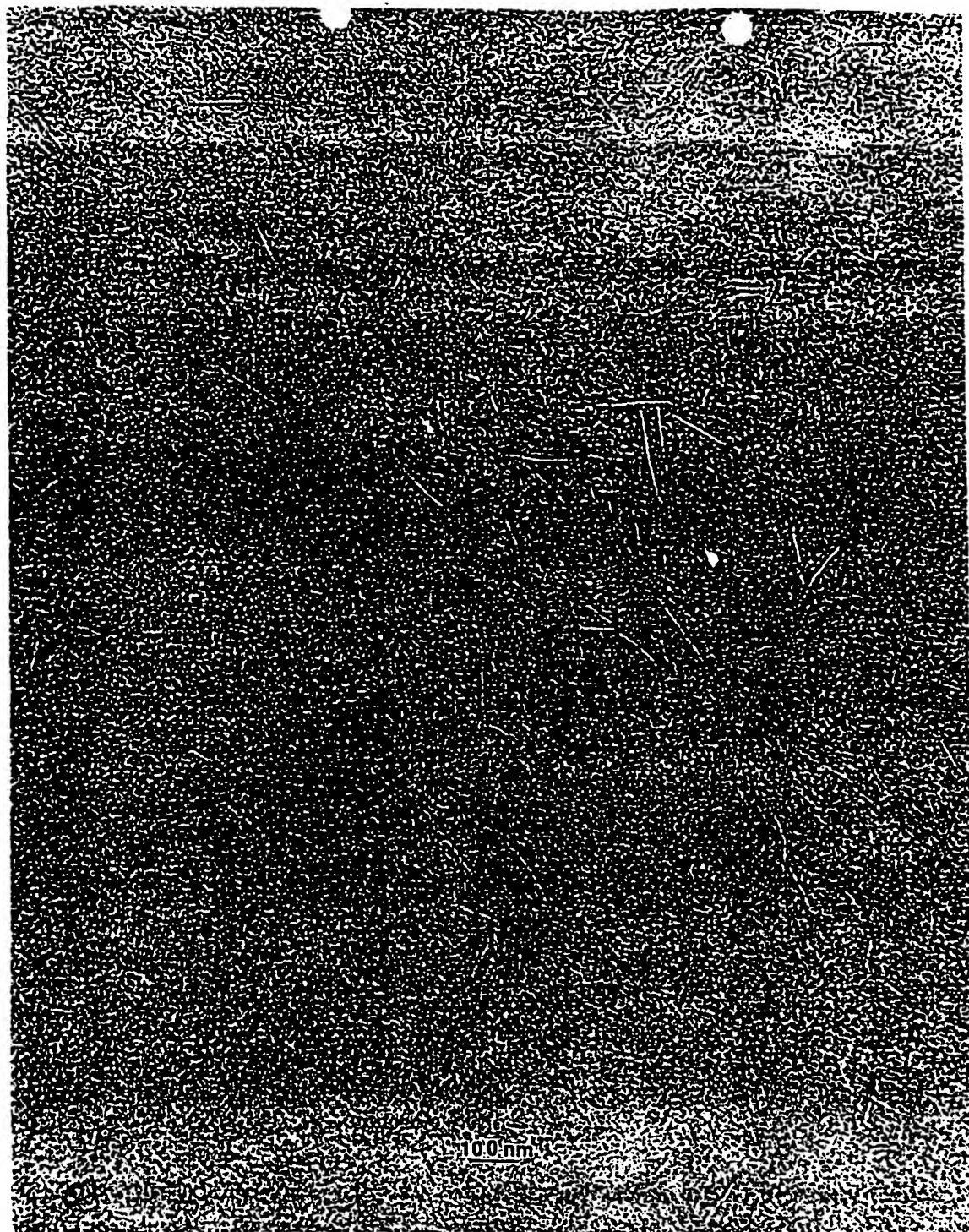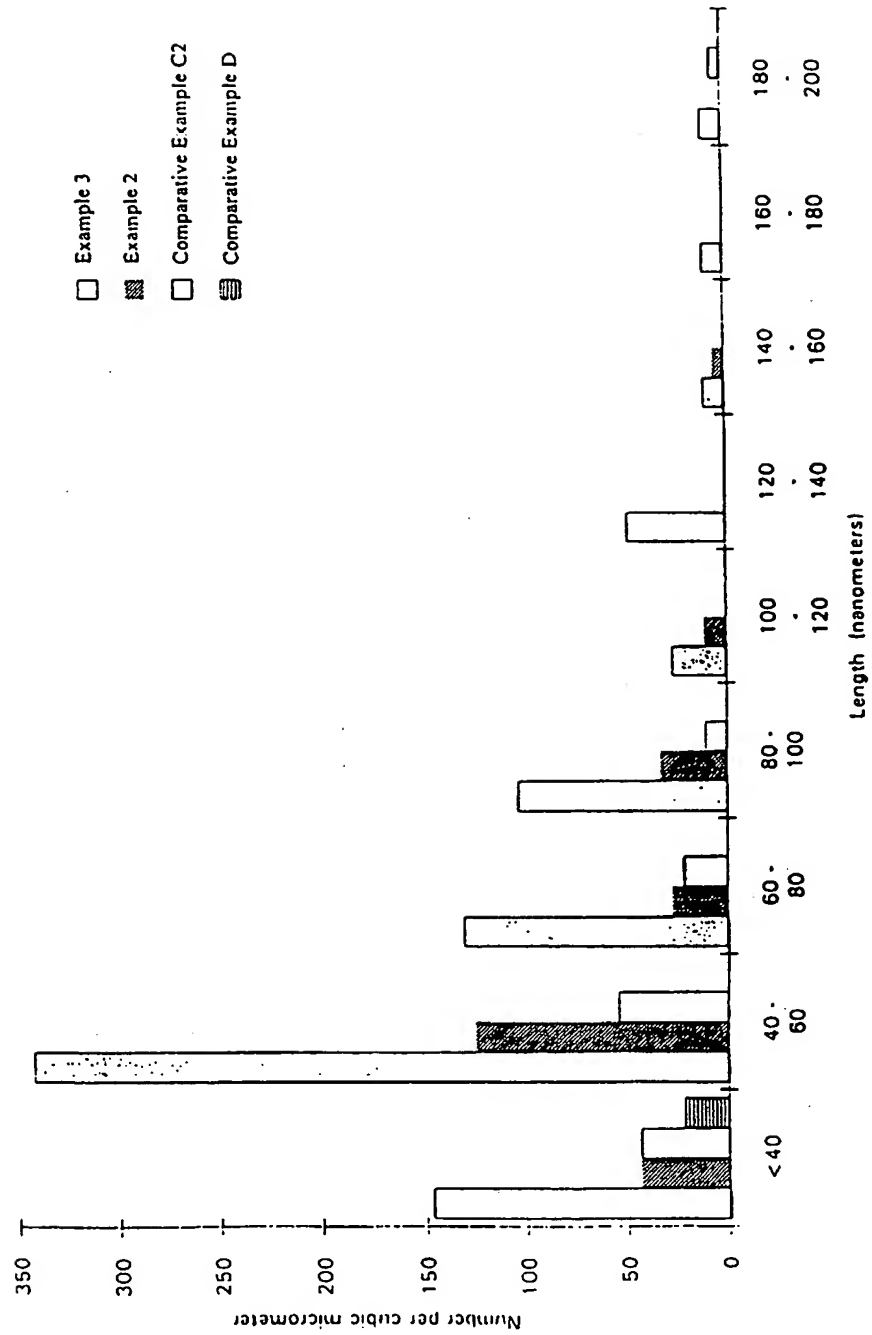
FIGURE 8

FIGURE 9

Figure 10
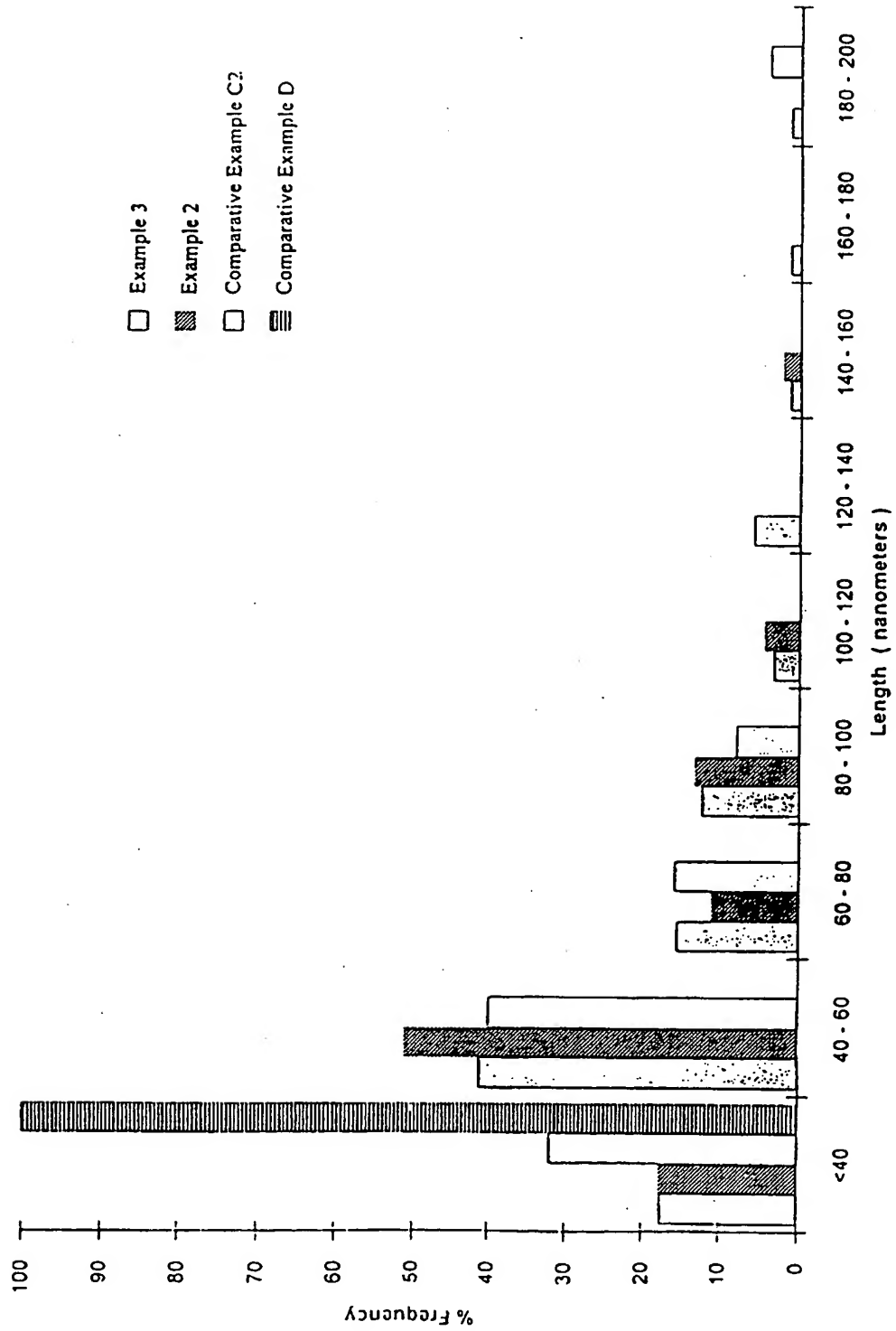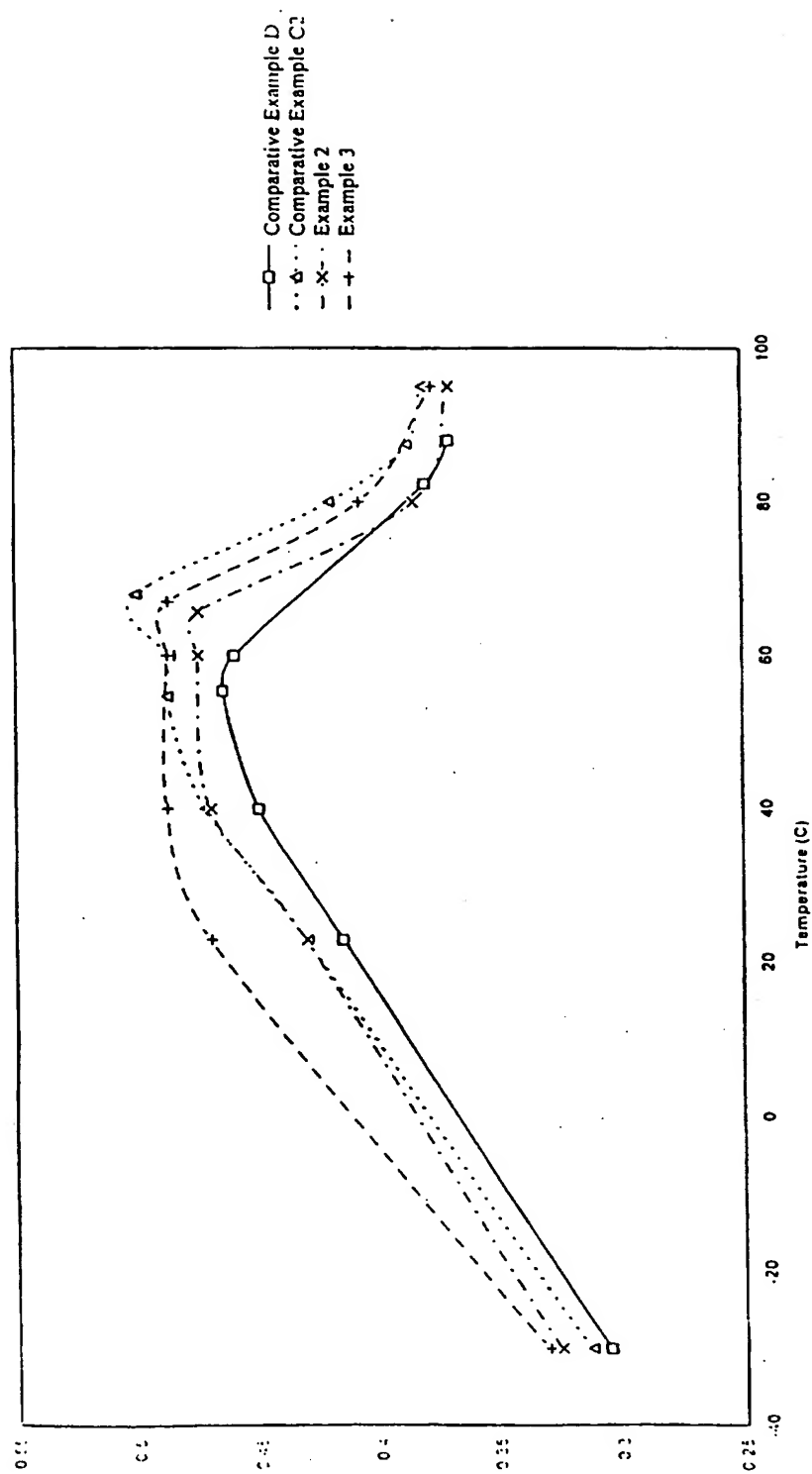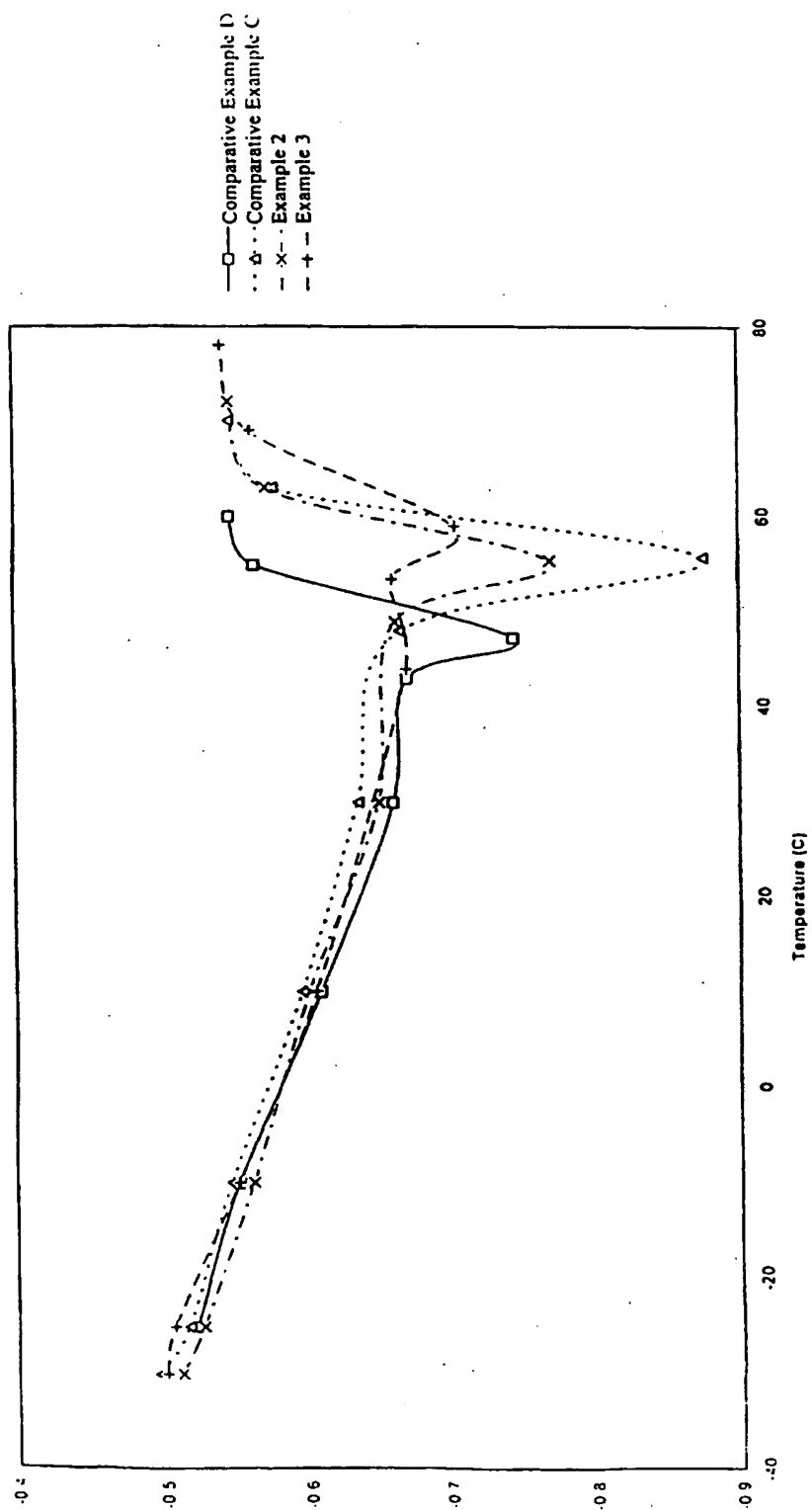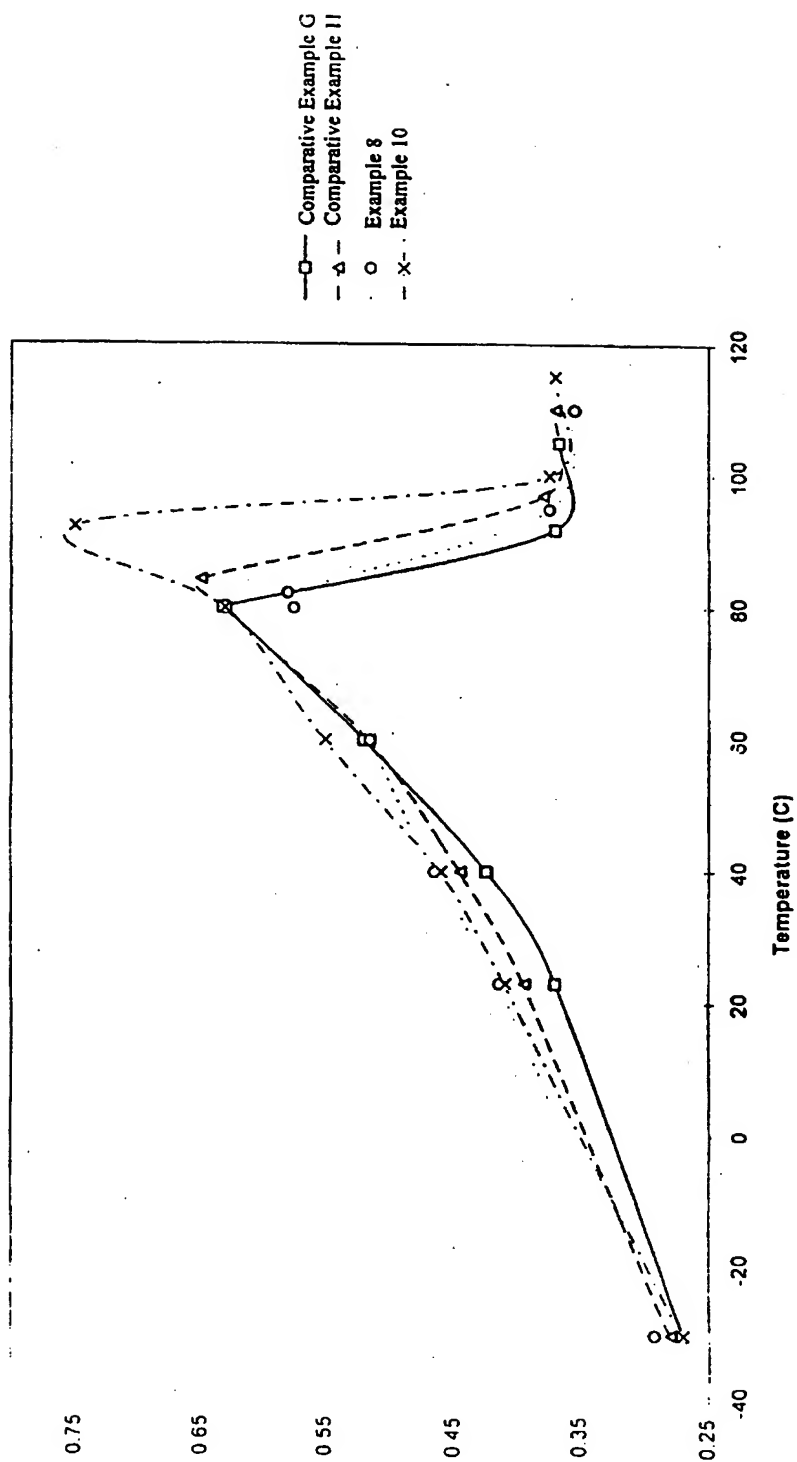
Figure 11

Figure 12

Figure 13
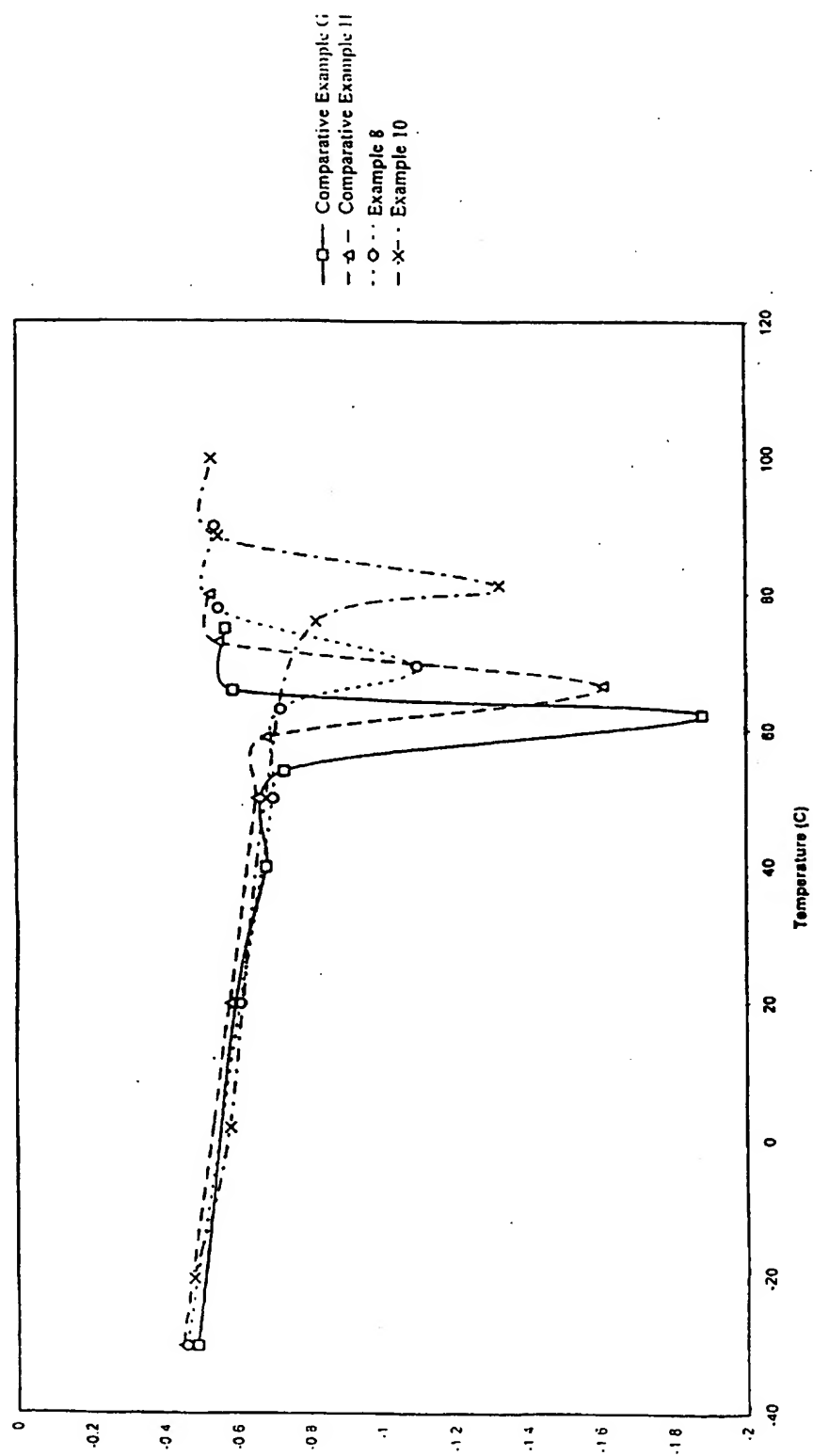
Figure 14

Figure 15
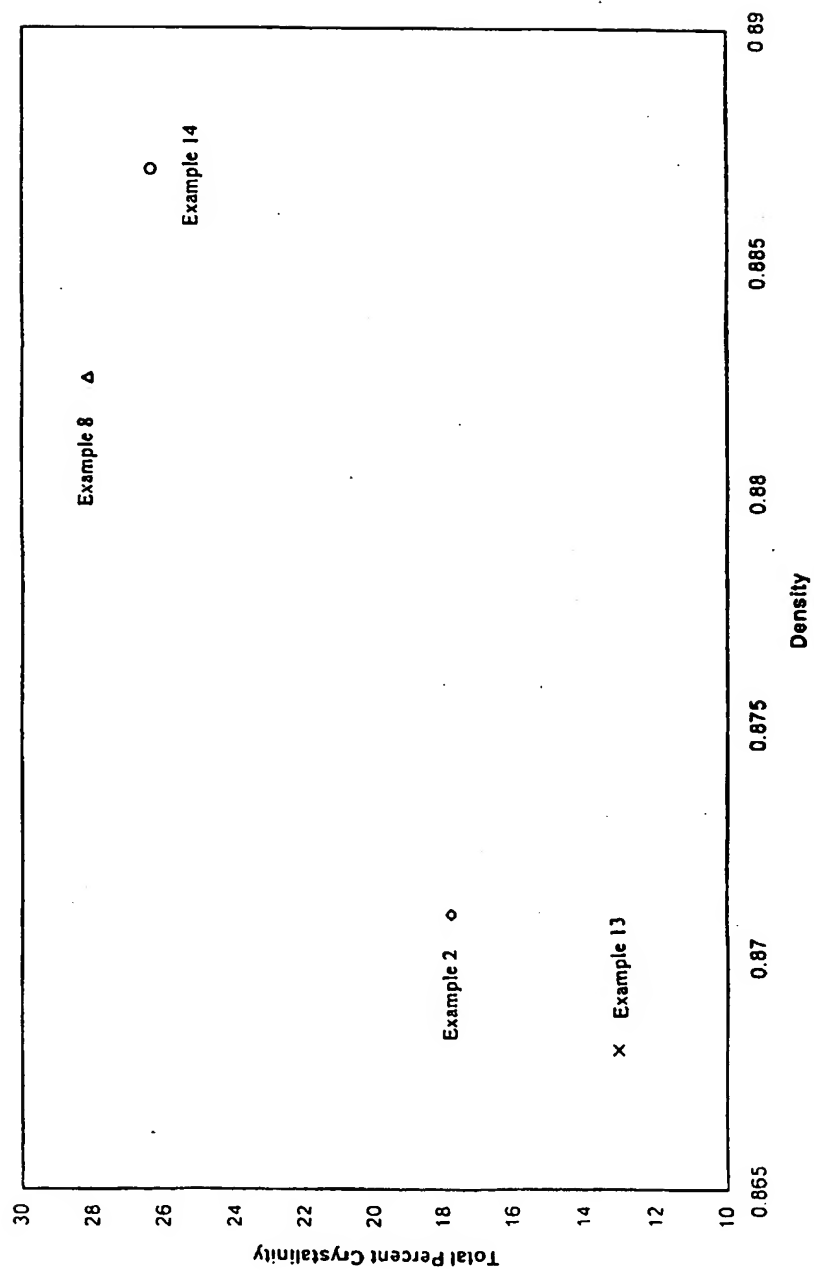
Figure 16
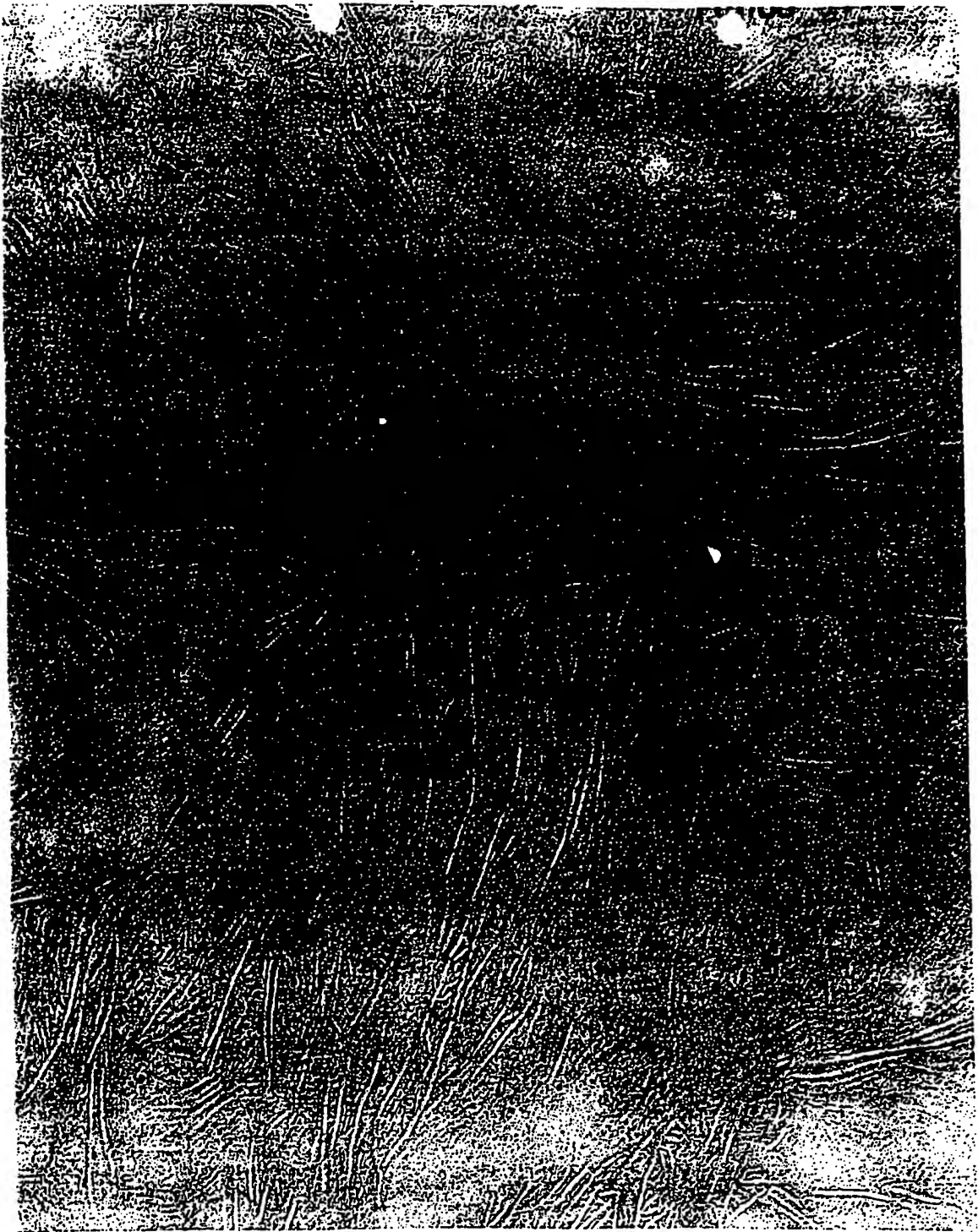
FIGURE 17

INTERNATIONAL SEARCH REPORT

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6   C08F210/02   C08F210/16

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6   C08F

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 435 250 A (UNION CARBIDE CHEM PLASTIC) 3 July 1991 see page 4, line 16 - line 21 see page 7, line 40 - line 48; claims; table 1 ---	1-6, 8-13,22
X	EP 0 321 851 A (HOECHST AG) 28 June 1989 see page 2, line 21 - line 25 see page 5, line 32 - line 37 see page 6, line 2 - line 10; claims ---	1-6,11, 12,22
X	EP 0 602 509 A (HOECHST AG) 22 June 1994 see page 9, line 54 - page 10, line 3; claims --- -/--	1-6

X	Further documents are listed in the continuation of box C.		X	Patent family members are listed in annex.

* Special categories of cited documents :

'A' document defining the general state of the art which is not considered to be of particular relevance

'E' earlier document but published on or after the international filing date

'L' document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

'O' document referring to an oral disclosure, use, exhibition or other means

'P' document published prior to the international filing date but later than the priority date claimed

'T' later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

'X' document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

'Y' document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

'&' document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
9 May 1997	26. 05. 97

Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+ 31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+ 31-70) 340-3016	Authorized officer Kaumann, E

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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1

Form PCT/ISA/210 (continuation of second sheet) (July 1992)

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